

Policy Assessment for the Review of the Secondary National Ambient Air Quality Standards for Oxides of Nitrogen, Oxides of Sulfur and Particulate Matter, External Review Draft

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U.S. Environmental Protection Agency Office of Air Quality Planning and Standards Health and Environmental Impacts Division Research Triangle Park, NC

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# **1 INTRODUCTION**

2 This document, Draft Policy Assessment for the Review of the Secondary National 3 Ambient Air Quality Standards for Oxides of Nitrogen, Oxides of Sulfur and Particulate Matter, 4 External Review Draft (hereafter referred to as draft PA), presents the draft policy assessment for 5 the U.S. Environmental Protection Agency's (EPA's) current review of the secondary national 6 ambient air quality standards (NAAQS) for oxides of nitrogen, oxides of sulfur and particulate matter (SOx and PM).<sup>12</sup> In the context of the secondary standards for oxides of nitrogen, oxides 7 of sulfur and PM, the scope pertains to the protection of the public welfare from adverse effects 8 9 related to ecological effects this draft PA considers key policy-relevant issues, drawing on those 10 identified in the Integrated Review Plan for the Secondary National Ambient Air Quality 11 Standards for Ecological Effects of Oxides of Nitrogen, Oxides of Sulfur and Particulate Matter 12 (IRP; U.S. EPA, 2017 and the Integrated Science Assessment for Oxides of Nitrogen, Oxides of 13 Sulfur and Particulate Matter – Ecological Criteria (ISA; [U.S. EPA, 2020]). 14 This document is organized into seven chapters, encompassing information on air quality, 15 the nature of effects and exposure conditions associated with effects, relationships between 16 deposition and air quality metrics, and a review of the standards. A detailed description of chapters within this document (and associated appendices) is provided in section 1.5 below. In 17 18 this introductory chapter, we present information on the purpose of the PA (section 1.1), 19 legislative requirements for reviews of the NAAQS (section 1.2), and an overview of the history 20 of the N oxides, SOx and PM NAAQS, including background information on prior reviews 21 (Section 1.3). Section 1.4 describes progress and next steps in the current review.

#### 22 **1.1 PURPOSE**

1

The PA, when final, presents an evaluation, for consideration by the EPA Administrator,
of the policy implications of the currently available scientific information, assessed in the ISA,

<sup>&</sup>lt;sup>1</sup> This review focuses on the presence in ambient air of oxides of nitrogen, oxides of sulfur, and particulate matter. The standards that are the focus of this review are the secondary standards for NO<sub>2</sub>, set in 1971 (36 FR 8186, April 30, 1971), for SO<sub>2</sub>, set in 1971 (36 FR 8186, April 30, 1971), for PM<sub>10</sub>, set in 2012 (78 FR 3085, January 15, 2013), and for PM<sub>2.5</sub>, set in 2012 (78 FR 3085, January 15, 2013). These standards are referred to in this document as the "current" or "existing" standards.

<sup>&</sup>lt;sup>2</sup> This review differs from the review of the secondary standards for oxides of nitrogen and sulfur completed in 2012 in that the current review includes consideration of the secondary PM standards, in addition to the secondary standards for oxides of nitrogen and sulfur. Given the contribution of nitrogen compounds to PM, including but not limited to those related to oxides of nitrogen, the current review provides for an expanded and more integrated consideration of N deposition and the current related air quality information. Regarding PM, welfare effects associated with visibility impairment, climate effects, and materials effects (i.e., damage and soiling) are being addressed in the separate review of the NAAQS for PM.

any quantitative air quality, exposure or risk analyses based on the ISA findings, and related
limitations and uncertainties.<sup>3</sup> Ultimately, final decisions on the secondary N oxides, SOx, and
PM NAAQS will reflect the judgments of the Administrator. The role of the PA is to help
"bridge the gap" between the Agency's scientific assessment and quantitative technical analyses,
and the judgments required of the Administrator in determining whether it is appropriate to retain
or revise the NAAQS.

In evaluating the question of adequacy of the current standards and whether it may be appropriate to consider alternative standards, the PA focuses on information that is most pertinent to evaluating the standards and their basic elements: indicator, averaging time, form, and level.<sup>4</sup> These elements, which together serve to define each standard, must be considered collectively in evaluating the public health and public welfare protection the standards afford. The development of the PA is also intended to facilitate advice to the Agency and recommendations to the Administrator from an independent scientific review committee, the

14 Clean Air Scientific Advisory Committee (CASAC), as provided for in the Clean Air Act

15 (CAA). As discussed below in section 1.2, the CASAC is to advise on subjects including the

16 Agency's assessment of the relevant scientific information and on the adequacy of the current

17 standards, and to make recommendations as to any revisions of the standards that may be

18 appropriate. The EPA generally makes available to the CASAC and the public one or more drafts

19 of the PA for CASAC review and public comment.

In this draft PA, we consider the available scientific information, as assessed in the
 *Integrated Science Assessment for Oxides of Nitrogen, Oxides of Sulfur and Particulate Matter – Ecological Criteria*, (ISA [U.S. EPA, 2020]) which included literature through May 2017, and

23 additional policy-relevant quantitative air quality, exposure and risk analyses. Advice and

24 comments from the CASAC and the public on this draft PA will inform the evaluation and

25 conclusions in the final PA.

The PA is designed to assist the Administrator in considering the currently available
scientific and risk information and formulating judgments regarding the standards. The final PA

28 will inform the Administrator's decision in this review. Beyond informing the Administrator and

<sup>&</sup>lt;sup>3</sup> The terms "staff," "we," and "our" throughout this document refer to the staff in the EPA's Office of Air Quality Planning and Standards (OAQPS).

<sup>&</sup>lt;sup>4</sup> The indicator defines the chemical species or mixture to be measured in the ambient air for the purpose of determining whether an area attains the standard. The averaging time defines the period over which air quality measurements are to be averaged or otherwise analyzed. The form of a standard defines the air quality statistic that is to be compared to the level of the standard in determining whether an area attains the standard. For example, the form of the annual NAAQS for fine particulate matter is the average of annual mean concentrations for three consecutive years, while the form of the 3-hour secondary NAAQS for SO<sub>2</sub> is the second-highest 3-hour average in a year. The level of the standard defines the air quality concentration used for that purpose.

facilitating the advice and recommendations of the CASAC, the PA is also intended to be a useful reference to all interested parties. In these roles, it is intended to serve as a source of policy-relevant information that supports the Agency's review of the secondary NAAQS for N oxides, SOx, and PM, and it is written to be understandable to a broad audience.

5 1

#### **1.2 LEGISLATIVE REQUIREMENTS**

6 Two sections of the Clean Air Act (CAA) govern the establishment and revision of the 7 NAAQS. Section 108 (42 U.S.C. 7408) directs the Administrator to identify and list certain air 8 pollutants and then to issue air quality criteria for those pollutants. The Administrator is to list 9 those pollutants "emissions of which, in his judgment, cause or contribute to air pollution which 10 may reasonably be anticipated to endanger public health or welfare"; "the presence of which in 11 the ambient air results from numerous or diverse mobile or stationary sources"; and for which he "plans to issue air quality criteria...." (42 U.S.C. § 7408(a)(1)). Air quality criteria are intended 12 13 to "accurately reflect the latest scientific knowledge useful in indicating the kind and extent of all 14 identifiable effects on public health or welfare which may be expected from the presence of [a] 15 pollutant in the ambient air...." 42 U.S.C. § 7408(a)(2).

16 Section 109 [42 U.S.C. 7409] directs the Administrator to propose and promulgate 17 "primary" and "secondary" NAAOS for pollutants for which air quality criteria are issued [42] 18 U.S.C. § 7409(a)]. Under section 109(b)(2), a secondary standard must "specify a level of air 19 quality the attainment and maintenance of which, in the judgment of the Administrator, based on 20 such criteria, is requisite to protect the public welfare from any known or anticipated adverse effects associated with the presence of [the] pollutant in the ambient air."<sup>5</sup> 21 22 In setting primary and secondary standards that are "requisite" to protect public health 23 and welfare, respectively, as provided in section 109(b), the EPA's task is to establish standards

that are neither more nor less stringent than necessary. In so doing, the EPA may not consider the

costs of implementing the standards. See generally, *Whitman v. American Trucking Ass'ns*, 531

26 U.S. 457, 465-472, 475-76 (2001). Likewise, "[a]ttainability and technological feasibility are not

27 relevant considerations in the promulgation of national ambient air quality standards" (American

28 Petroleum Institute v. Costle, 665 F.2d 1176, 1185 [D.C. Cir. 1981]). At the same time, courts

- 29 have clarified the EPA may consider "relative proximity to peak background ... concentrations"
- 30 as a factor in deciding how to revise the NAAQS in the context of considering standard levels

<sup>&</sup>lt;sup>5</sup> Under CAA section 302(h) (42 U.S.C. § 7602(h)), effects on welfare include, but are not limited to, "effects on soils, water, crops, vegetation, manmade materials, animals, wildlife, weather, visibility, and climate, damage to and deterioration of property, and hazards to transportation, as well as effects on economic values and on personal comfort and well-being."

1 within the range of reasonable values supported by the air quality criteria and judgments of the 2 Administrator (American Trucking Ass'ns, v. EPA, 283 F.3d 355, 379 [D.C. Cir. 2002]). 3 Section 109(d)(1) of the Act requires periodic review and, if appropriate, revision of 4 existing air quality criteria to reflect advances in scientific knowledge on the effects of the 5 pollutant on public health and welfare. Under the same provision, the EPA is also to periodically 6 review and, if appropriate, revise the NAAQS, based on the revised air quality criteria.<sup>6</sup> 7 Section 109(d)(2) addresses the appointment and advisory functions of an independent 8 scientific review committee. Section 109(d)(2)(A) requires the Administrator to appoint this 9 committee, which is to be composed of "seven members including at least one member of the 10 National Academy of Sciences, one physician, and one person representing State air pollution 11 control agencies." Section 109(d)(2)(B) provides that the independent scientific review 12 committee "shall complete a review of the criteria...and the national primary and secondary 13 ambient air quality standards...and shall recommend to the Administrator any new...standards 14 and revisions of existing criteria and standards as may be appropriate...." Since the early 1980s, 15 this independent review function has been performed by the Clean Air Scientific Advisory 16 Committee (CASAC) of the EPA's Science Advisory Board.

# 17 1.3 BACKGROUND ON CRITERIA AND SECONDARY STANDARDS 18 FOR NITROGEN AND SULFUR OXIDES AND PARTICULATE 19 MATTER

Secondary NAAQS were first established for oxides of nitrogen and oxides of sulfur in
1971 (36 FR 8186, April 30, 1971) based on evidence available regarding their effects on
vegetation. The secondary NAAQS for PM were first established in 1971 (36 FR 8186, April
30,1971). Since that time, the EPA has periodically reviewed the air quality criteria and
standards, with the most recent review being completed in 2012. The details of these reviews are
described in the subsections below.

#### 26 1.3.1 Nitrogen Oxides

The EPA first promulgated identical primary and secondary NAAQS for nitrogen dioxide (NO<sub>2</sub>) in April 1971 after reviewing the relevant science on the public health and welfare effects associated with oxides of nitrogen in the 1971 Air Quality Criteria Document (AQCD). These standards were set at a level of 0.053 parts per million (ppm) as an annual average (36 FR 8186, April 30, 1971). In 1982, the EPA published Air Quality Criteria for Oxides of Nitrogen (U.S. EPA, 1982), which updated the scientific criteria upon which the initial standards were based. In

<sup>&</sup>lt;sup>6</sup> This section of the Act requires the Administrator to complete these reviews and make any revisions that may be appropriate "at five-year intervals."

1 February 1984, the EPA proposed to retain these standards (49 FR 6866, February 23, 1984).

- After considering public comments, the EPA published the final decision to retain these
  standards in June 1985 (50 FR 25532, June 19, 1985).
- 4 The EPA began a second review of the oxides of nitrogen secondary standards in 1987. 5 In November 1991 the EPA released an updated AQCD for CASAC and public review and 6 comment (56 FR 59285, November 25, 1991), which provided a comprehensive assessment of 7 the available scientific and technical information on health and welfare effects associated with 8 NO<sub>2</sub> and other oxides of nitrogen. The CASAC reviewed the draft document at a meeting held on 9 July 1, 1993 and concluded in a closure letter to the Administrator that the document "provides a 10 scientifically balanced and defensible summary of current knowledge of the effects of this 11 pollutant and provides an adequate basis for the EPA to make a decision as to the appropriate 12 NAAQS for NO<sub>2</sub>" (Wolff, 1993). The Air Quality Criteria for Oxides of Nitrogen was then 13 finalized (U.S. EPA, 1993). The EPA's Office of Air Quality Planning and Standards (OAQPS) 14 also prepared a Staff Paper that summarized and integrated the key studies and scientific 15 evidence contained in the revised AQCD for oxides of nitrogen and identified the critical 16 elements to be considered in the review of the NO<sub>2</sub> NAAQS. CASAC reviewed two drafts of the 17 Staff Paper and concluded in a closure letter to the Administrator that the document provided a 18 "scientifically adequate basis for regulatory decisions on nitrogen dioxide" (Wolff, 1995). 19 In October 1995 the Administrator announced her proposed decision not to revise the 20 secondary NAAQS for NO<sub>2</sub> (60 FR 52874; October 11, 1995). A year later, the Administrator 21 made a final determination not to revise the NAAQS for NO<sub>2</sub> after careful evaluation of the 22 comments received on the proposal (61 FR 52852; October 8, 1996). The secondary NAAQS for 23 NO<sub>2</sub> remains 0.053 ppm (100 micrograms per cubic meter  $[\mu g/m^3]$  of air), annual arithmetic 24 average, calculated as the arithmetic mean of the 1-hour NO<sub>2</sub> concentrations.

#### 25 **1.3.2 Sulfur Oxides**

26 The EPA first promulgated secondary NAAQS for sulfur dioxide (SO<sub>2</sub>) in April 1971 (36 27 FR 8186, April 30, 1971). The 1971 secondary standards for SO<sub>2</sub> were established solely on the 28 basis of evidence of adverse effects on vegetation available in the 1969 AQCD (U.S. DHEW, 29 1969a [1969 AQCD]). The secondary standards included a standard set at 0.02 ppm, annual 30 arithmetic mean, and a 3- hour average standard set at 0.5 ppm, not to be exceeded more than 31 once per year. In 1973, revisions made to Chapter 5 ("Effects of Sulfur Oxide in the Atmosphere 32 on Vegetation") of Air Quality Criteria for Sulfur Oxides (U.S. EPA, 1973) indicated that it 33 could not properly be concluded that the vegetation injury reported resulted from the average 34 SO<sub>2</sub> exposure over the growing season, rather than from short-term peak concentrations. 35 Therefore, the EPA proposed (38 FR 11355, May 7, 1973) and then finalized (38 FR 25678,

September 14, 1973) a revocation of the annual mean secondary standard. At that time, the EPA
 was aware that then-current concentrations of oxides of sulfur in the ambient air had other public
 welfare effects, including effects on materials, visibility, soils, and water. However, the available

4 data were considered insufficient to establish a quantitative relationship between specific

5 ambient concentrations of oxides of sulfur and such public welfare effects (38 FR 25679,

6 September 14, 1973).

7 In 1979, the EPA announced that it was revising the AQCD for oxides of sulfur 8 concurrently with that for PM and would produce a combined PM and oxides of sulfur criteria 9 document. Following its review of a draft revised criteria document in August 1980, CASAC 10 concluded that acid deposition was a topic of extreme scientific complexity because of the 11 difficulty in establishing firm quantitative relationships among (1) emissions of relevant 12 pollutants (e.g.,  $SO_2$  and oxides of nitrogen), (2) formation of acidic wet and dry deposition 13 products, and (3) effects on terrestrial and aquatic ecosystems. CASAC also noted that acid 14 deposition involves, at a minimum, several different criteria pollutants: oxides of sulfur, oxides 15 of nitrogen, and the fine particulate fraction of suspended particles. CASAC felt that any 16 document on this subject should address both wet and dry deposition, since dry deposition was 17 believed to account for a substantial portion of the total acid deposition problem.

18 For these reasons, CASAC recommended that a separate, comprehensive document on 19 acid deposition be prepared prior to any consideration of using the NAAQS as a regulatory 20 mechanism for the control of acid deposition. CASAC also suggested that a discussion of acid 21 deposition be included in the AQCDs for oxides of nitrogen and PM and oxides of sulfur. 22 Following CASAC closure on the AQCD for oxides of sulfur in December 1981, the EPA's 23 OAQPS published a Staff Paper in November 1982 (U.S. EPA, 1982), although the paper did not 24 directly assess the issue of acid deposition. Instead, the EPA subsequently prepared the following documents to address acid deposition: The Acidic Deposition Phenomenon and Its Effects: 25 26 Critical Assessment Review Papers, Volumes I and II (U.S. EPA, 1984a, b) and The Acidic 27 Deposition Phenomenon and Its Effects: Critical Assessment Document (U.S. EPA, 1985) (53 28 FR 14935 -14936, April 26, 1988). These documents, though they were not considered criteria 29 documents and did not undergo CASAC review, represented the most comprehensive summary 30 of scientific information relevant to acid deposition completed by the EPA at that point. 31 In April 1988 (53 FR 14926, April 26, 1988), the EPA proposed not to revise the existing 32 secondary standards for SO<sub>2</sub>. This proposed decision with regard to the secondary SO<sub>2</sub> NAAQS 33 was due to the Administrator's conclusions that (1) based upon the then-current scientific 34 understanding of the acid deposition problem, it would be premature and unwise to prescribe any 35 regulatory control program at that time and (2) when the fundamental scientific uncertainties had

1 been decreased through ongoing research efforts, the EPA would draft and support an

2 appropriate set of control measures.

#### 3 1.3.3 Particulate Matter

The EPA first established NAAQS for PM in 1971 (36 FR 8186, April 30, 1971), based on the original AQCD (U.S. DHEW, 1969b) and recognition of effects on vegetation and to match the primary standards that were set concurrently to protect human health.7 The secondary standards were set at 150 μg/m3, 24-hour average, from total suspended particles (TSP), not to be exceeded more than once per year, and 60 μg/m3, annual geometric mean.

In October 1979 (44 FR 56730, October 2, 1979), the EPA announced the first periodic
review of the air quality criteria and NAAQS for PM. Revised secondary standards were
promulgated in 1987 (52 FR 24634, July 1, 1987). In the 1987 decision, the EPA changed the
indicator for PM from TSP to PM10, and the level of the 24-hour secondary standard was set at
150 µg/m3, and the form was one expected exceedance per year, on average over three years.
The level of the annual secondary standard was set at 50 µg/m3, and the form was annual

- 15 arithmetic mean, averaged over three years.
- 16 In April 1994, the EPA announced its plans for the second periodic review of the air
- 17 quality criteria and NAAQS for PM, and in 1997 the EPA promulgated revisions to the NAAQS

18 (62 FR 38652, July 18, 1997). In the 1997 decision, the EPA determined that the fine and coarse

- 19 fractions of PM10 should be considered separately. The EPA added new standards, using PM2.5
- 20 as the indicator for fine particles (with PM2.5 referring to particles with a nominal mean
- 21  $\,$  aerodynamic diameter less than or equal to 2.5  $\mu m$  ). The EPA revised the secondary standards by
- 22 setting them equal in all respects to the primary standards as follows: (1) an annual standard with
- 23 a level of 15.0  $\mu$ g/m3, based on the 3-year average of annual arithmetic mean PM2.5
- 24 concentrations from single or multiple community-oriented monitors;8 and (2) a 24-hour
- standard with a level of 65  $\mu$ g/m3, based on the 3-year average of the 98th percentile of 24-hour
- 26 PM2.5 concentrations at each monitor within an area. Also, the EPA established a new reference
- 27 method for the measurement of PM2.5 in the ambient air and adopted rules for determining

<sup>&</sup>lt;sup>7</sup> Prior to the review initiated in 2007 (see below), the AQCD provided the scientific foundation (i.e., the air quality criteria) for the NAAQS. Beginning in that review, the Integrated Science Assessment (ISA) has replaced the AQCD.

<sup>&</sup>lt;sup>8</sup> The 1997 annual PM<sub>2.5</sub> standard was compared with measurements made at the community-oriented monitoring site recording the highest concentration or, if specific constraints were met, measurements from multiple community-oriented monitoring sites could be averaged (i.e., spatial averaging"). In the last review (completed in 2012) the EPA replaced the term "community-oriented" monitor with the term "area-wide" monitor. Area-wide monitors are those sited at the neighborhood scale or larger, as well as those monitors sited at micro- or middle-scales that are representative of many such locations in the same core-based statistical area (CBSA) (78 FR 3236, January 15, 2013).

1 attainment of the new standards. To continue to address the health effects of the coarse fraction

2 of PM10 (referred to as thoracic coarse particles or PM10-2.5; generally including particles with

3 a nominal mean aerodynamic diameter greater than 2.5  $\mu$ m and less than or equal to 10  $\mu$ m), the

4 EPA retained the primary annual PM10 standard and revised the form of the primary 24-hour

5 PM10 standard to be based on the 99th percentile of 24-hour PM10 concentrations at each

6 monitor in an area.

7 Following promulgation of the 1997 PM NAAQS, petitions for review were filed by 8 several parties, addressing a broad range of issues. In May 1999, the U.S. Court of Appeals for 9 the District of Columbia Circuit (D.C. Circuit) upheld the EPA's decision to establish fine 10 particle standards, holding that "the growing empirical evidence demonstrating a relationship 11 between fine particle pollution and adverse health effects amply justifies establishment of new 12 fine particle standards." American Trucking Associations, Inc. v. EPA, 175 F. 3d 1027, 1055-56 13 (D.C. Cir. 1999). The D.C. Circuit also found "ample support" for the EPA's decision to regulate 14 coarse particle pollution, but vacated the 1997 PM10 standards, concluding that the EPA had not 15 provided a reasonable explanation justifying use of PM10 as an indicator for coarse particles. 16 American Trucking Associations v. EPA, 175 F. 3d at 1054-55. Pursuant to the D.C. Circuit's 17 decision, the EPA removed the vacated 1997 PM10 standards, and the pre-existing 1987 PM10 18 standards remained in place (65 FR 80776, December 22, 2000). The D.C. Circuit also upheld 19 the EPA's determination not to establish more stringent secondary standards for fine particles to 20 address effects on visibility (American Trucking Associations v. EPA, 175 F. 3d at 1027).

The D.C. Circuit also addressed more general issues related to the NAAQS, including issues related to the consideration of costs in setting NAAQS and the EPA's approach to establishing the levels of NAAQS. Regarding the cost issue, the court reaffirmed prior rulings holding that in setting NAAQS the EPA is "not permitted to consider the cost of implementing

those standards." See generally, Whitman v. American Trucking Ass'ns, 531 U.S. 457, 465-472,

26 475-76 (2001). Likewise, "[a]ttainability and technological feasibility are not relevant

27 considerations in the promulgation of national ambient air quality standards" (American

28 Petroleum Institute v. Costle, 665 F.2d 1176, 1185 [D.C. Cir. 1981], cert. denied, 455 U.S. 1034

29 [1982]; accord Murray Energy Corp. v. EPA, 936 F.3d 597, 623-24 [D.C. Cir. 2019]). At the

30 same time, courts have clarified the EPA may consider "relative proximity to peak background

31 ... concentrations" as a factor in deciding how to revise the NAAQS in the context of

32 considering standard levels within the range of reasonable values supported by the air quality

33 criteria and judgments of the Administrator (American Trucking Ass'ns, v. EPA, 283 F.3d 355,

34 379 [D.C. Cir. 2002], hereafter referred to as "ATA III").

In October 1997, the EPA published its plans for the third periodic review of the air
 quality criteria and NAAQS for PM (62 FR 55201, October 23, 1997). After the CASAC and

1 public review of several drafts, the EPA's National Center for Environmental Assessment 2 finalized the AQCD in October 2004 (U.S. EPA, 2004a and 2004b). The EPA's Office of Air 3 Quality Planning and Standards (OAQPS) finalized a Risk Assessment and Staff Paper in 4 December 2005 (Abt Associates, 2005, U.S. EPA, 2005).9 On December 20, 2005, the EPA 5 announced its proposed decision to revise the NAAQS for PM and solicited public comment on a 6 broad range of options (71 FR 2620, January 17, 2006). On September 21, 2006, the EPA 7 announced its final decisions to revise the primary and secondary NAAQS for PM to provide 8 increased protection of public health and welfare, respectively (71 FR 61144, October 17, 2006). 9 With regard to the primary and secondary standards for fine particles, the EPA revised the level 10 of the 24-hour PM2.5 standards to 35  $\mu$ g/m3, retained the level of the annual PM2.5 standards at 11 15.0 µg/m3, and revised the form of the annual PM2.5 standards by narrowing the constraints on 12 the optional use of spatial averaging. With regard to the primary and secondary standards for 13 PM10, the EPA retained the 24-hour standards, with levels at 150  $\mu$ g/m3, and revoked the annual 14 standards.10 The Administrator judged that the available evidence generally did not suggest a 15 link between long-term exposure to existing ambient levels of coarse particles and health or 16 welfare effects. In addition, a new reference method was added for the measurement of 17 PM10-2.5 in the ambient air in order to provide a basis for approving federal equivalent methods 18 (FEMs) and to promote the gathering of scientific data to support future reviews of the PM 19 NAAQS. 20 Several parties filed petitions for review following promulgation of the revised PM 21 NAAQS in 2006. One of these petitions addressed the issue of setting the secondary PM2.5 22 standards identical to the primary standards. On February 24, 2009, the D.C. Circuit issued its

opinion in the case American Farm Bureau Federation v. EPA, 559 F. 3d 512 (D.C. Cir. 2009)
 and remanded the standards to the EPA because the Agency failed to adequately explain why

- and remanded the standards to the EPA because the Agency failed to adequately explain why
   setting the secondary PM standards identical to the primary standards provided the required
- 26 protection for public welfare, including protection from visibility impairment. Id. at 528-32. The

<sup>&</sup>lt;sup>9</sup> Prior to the review initiated in 2007, the Staff Paper presented the EPA staff's considerations and conclusions regarding the adequacy of existing NAAQS and, when appropriate, the potential alternative standards that could be supported by the evidence and information. More recent reviews present this information in the Policy Assessment.

<sup>&</sup>lt;sup>10</sup> In the 2006 proposal, the EPA proposed to revise the 24-hour PM<sub>10</sub> standard in part by establishing a new PM<sub>10-2.5</sub> indicator for thoracic coarse particles (i.e., particles generally between 2.5 and 10 µm in diameter). The EPA proposed to include any ambient mix of PM<sub>10-2.5</sub> that was dominated by resuspended dust from high density traffic on paved roads and by PM from industrial sources and construction sources. The EPA proposed to exclude any ambient mix of PM<sub>10-2.5</sub> that was dominated by rural windblown dust and soils and by PM generated from agricultural and mining sources. In the final decision, the existing PM<sub>10</sub> standard was retained, in part due to an "inability…to effectively and precisely identify which ambient mixes are included in the [PM<sub>10-2.5</sub>] indicator and which are not" (71 FR 61197, October 17, 2006).

1 EPA responded to the court's remands as part of the next review of the PM NAAQS, which was 2 initiated in 2007.

3 In June 2007, the EPA initiated the fourth periodic review of the air quality criteria and 4 the PM NAAQS by issuing a call for information in the Federal Register (72 FR 35462, June 28, 5 2007). Based on the NAAQS review process, as revised in 2008 and again in 2009, the EPA held 6 science/policy issue workshops on the primary and secondary PM NAAQS (72 FR 34003, June 7 20, 2007; 72 FR 34005, June 20, 2007), and prepared and released the planning and assessment 8 documents that comprise the review process (i.e., IRP (U.S. EPA, 2008b), ISA (U.S. EPA, 9 2009b), Risk and Exposure Assessment (REA) planning document for welfare (U.S. EPA, 10 2009c), and an urban-focused visibility assessment (U.S. EPA, 2010), and PA (U.S. EPA, 2011). 11 In June 2012, the EPA announced its proposed decision to revise the NAAQS for PM (77 FR 12 38890, June 29, 2012). In December 2012, the EPA announced its final decisions to revise only 13 the primary NAAQS for PM to provide increased protection of public health (78 FR 3086, 14 January 15, 2013). The PM secondary standards were established to provide protection against a

15 variety of PM-associated welfare effects, including effects on vegetation as well as visibility

16 impairment and materials damage (e.g., soiling, corrosion). The EPA generally retained the 24-

17 hour and annual PM2.5 standards, set at 35  $\mu$ g/m and 15  $\mu$ g/m and the 24-hour PM10 standard,

18 set at a level of 150  $\mu$ g/m3, to address visibility and non-visibility welfare effects.

#### 19 **1.3.4** Last Review of the Criteria and Secondary Standards for Nitrogen and Sulfur 20 Oxides

21 The EPA initiated the prior review in December 2005, with a call for information (70 FR 22 73236) for the development of a revised ISA. An Integrated Review Plan (IRP) was developed to 23 provide the framework and schedule as well as the scope of the review and to identify policy-24 relevant questions to be addressed in the components of the review. The IRP was released in 25 2007 (U.S. EPA, 2007) for CASAC and public review. The EPA held a workshop in July 2007 26 on the ISA to obtain broad input from the relevant scientific communities. This workshop helped 27 to inform the preparation of the first draft ISA, which was released for CASAC and public 28 review in December 2007; a CASAC meeting was held on April 2–3, 2008, to review the first 29 draft ISA. A second draft ISA was released for CASAC and public review in August 2008 and 30 was discussed at a CASAC meeting held on October 1–2, 2008. The final ISA (U.S. EPA, 31 2008a) was released in December 2008. 32 Based on the science presented in the ISA, the EPA developed the REA to further assess

33 the national impact of the effects documented in the ISA. The Draft Scope and Methods Plan for

34

Risk/ Exposure Assessment: Secondary NAAOS Review for Oxides of Nitrogen and Oxides of

35 Sulfur outlining the scope and design of the future REA was prepared for CASAC consultation 1 and public review in March 2008. A first draft REA was presented to CASAC and the public for

- 2 review in August 2008, and a second draft was presented for review in June 2009. The final REA
- 3 (U.S. EPA, 2009a) was released in September 2009.
- 4 A first draft PA was released in March 2010, and reviewed by CASAC on April 1–2, 5 2010. In a June 22, 2010, letter to the Administrator, CASAC provided advice and 6 recommendations to the Agency concerning the first draft PA (Russell and Samet, 2010a). A 7 second draft PA was released to CASAC and the public in September 2010, and reviewed by 8 CASAC on October 6–7, 2010. The CASAC provided advice and recommendations to the 9 Agency regarding the second draft PA in a December 9, 2010 letter (Russell and Samet 2010b). 10 The CASAC and public comments on the second draft PA were considered by the EPA staff in 11 developing a final PA (U.S. EPA, 2011). CASAC requested an additional meeting to provide 12 additional advice to the Administrator based on the final PA on February 15–16, 2011. On 13 January 14, 2011, the EPA released a version of the final PA prior to final document production, 14 to provide sufficient time for CASAC review of the document in advance of this meeting. The 15 final PA, incorporating final reference checks and document formatting, was released in 16 February 2011. In a May 17, 2011, letter (Russell and Samet, 2011), CASAC offered additional 17 advice and recommendations to the Administrator with regard to the review of the secondary 18 NAAQS for oxides of nitrogen and oxides of sulfur. 19 On August 1, 2011, the EPA published a proposed decision to retain the existing annual
- 20 average NO<sub>2</sub> and 3-hour average SO<sub>2</sub> secondary standards, recognizing the protection they 21 provided from direct effects on vegetation (76 FR 46084, August 1, 2011). In the proposal, the 22 Administrator further concluded that the existing NO<sub>2</sub> and SO<sub>2</sub> secondary standards were not 23 adequate to protect against the adverse impacts of acidification of both aquatic and terrestrial 24 ecosystems or nutrient enrichment of terrestrial ecosystems, and proposed to revise the 25 secondary standards by adding secondary standards identical to the NO<sub>2</sub> and SO<sub>2</sub> primary 1-hour 26 standards set in 2010, noting that these new standards<sup>11</sup> would result in reductions in oxides of 27 nitrogen and sulfur that would likely reduce nitrogen and sulfur deposition to sensitive 28 ecosystems (76 FR 46084, August 1, 2011).
- After consideration of public comments, the Administrator's final decision retained the existing standards to address the direct effects on vegetation of exposure to gaseous oxides of nitrogen and sulfur and did not set additional standards at that time to address effects associated with deposition of oxides of nitrogen and sulfur on sensitive aquatic and terrestrial ecosystems (77 FR 20218, April 3, 2012). The limitations and uncertainties in the available information were

<sup>&</sup>lt;sup>11</sup> The 2010 primary 1-hour standards include the NO<sub>2</sub> standard set at a level of 100 parts per billion (ppb) and the SO2 standard set at a level of 75 ppb.

1 judged to be too great to support establishment of a new standard that could be concluded to

- 2 provide the requisite protection for such effects under the Act. The Administrator concluded that
- 3 while the current secondary standards were not adequate to provide protection against potentially
- 4 adverse deposition-related effects associated with oxides of nitrogen and sulfur, it was not
- 5 appropriate under Section 109 to set any new secondary standards for such effects at that time.
- 6 The Administrator also determined that setting new secondary standards identical to the 7 existing 1-hour NO<sub>2</sub> and SO<sub>2</sub> primary standards would be neither necessary nor appropriate as, in 8 her judgment, such standards could not reasonably be judged to provide requisite protection of 9 public welfare. In addition, the Administrator decided that it was appropriate to retain the 10 existing NO<sub>2</sub> and SO<sub>2</sub> secondary standards to address direct effects of gaseous NO<sub>2</sub> and SO<sub>2</sub> on 11 vegetation. Thus, taken together, the Administrator decided to retain and not revise the current 12 NO<sub>2</sub> and SO<sub>2</sub> secondary standards: a NO<sub>2</sub> standard set at a level of 0.053 ppm, as an annual 13 arithmetic average, and a SO<sub>2</sub> standard set at a level of 0.5 ppm, as a 3-hour average, not to be
- arithmetic average, and a SO<sub>2</sub> standard set at a level of 0.5 ppm, as a 3-hour average, not to be
  exceeded more than once per year (77 FR 20281, April 3, 2012).

15 The EPA's 2012 decision was challenged by the Center for Biological Diversity and 16 other environmental groups. The petitioners argued that having decided that the existing 17 standards were not adequate to protect against adverse public welfare effects such as damage to 18 sensitive ecosystems; the Administrator was required to identify the requisite level of protection 19 for the public welfare and to issue a NAAQS to achieve and maintain that level of protection. 20 The D.C. Circuit disagreed, finding that the EPA acted appropriately in not setting a secondary 21 standard given the EPA's conclusions that "the available information was insufficient to permit a 22 reasoned judgment about whether any proposed standard would be 'requisite to protect the public welfare . . . '."<sup>12</sup> In reaching this decision, the court noted that the EPA had "explained in 23 24 great detail" the profound uncertainties associated with setting a secondary NAAOS to protect against aquatic acidification<sup>13</sup>. 25

- 26 **1.4 CURRENT REVIEW**
- 27 In August 2013, the EPA's National Center for Environmental Assessment (NCEA)
- 28 issued a call for information in the *Federal Register* for information related to the newly initiated
- 29 review of the air quality criteria for oxides of sulfur and oxides of nitrogen (78 FR 53452,
- 30 August 29, 2013). Two types of information were called for: information regarding significant

31 new research studies to be considered for the ISA for the review, and policy-relevant issues for

32 consideration in this NAAQS review. Based in part on the information received in response to

<sup>&</sup>lt;sup>12</sup> Center for Biological Diversity, et al. v. EPA, 749 F.3d 1079, 1087 (2014).

<sup>&</sup>lt;sup>13</sup> Id. at 1088.

- 1 the call for information, the EPA developed a draft IRP which was made available for
- 2 consultation with the CASAC and for public comment (80 FR 69220, November 9, 2015). In
- 3 developing the final IRP, the EPA expanded the review to include the ecological effects of PM.
- 4 Comments from the CASAC (Diez Roux and Fernandez, 2016) and the public on the draft IRP
- 5 were considered in preparing the final IRP (U.S. EPA, 2017).
- 6 In March 2017, the EPA released the first external review draft of the *Integrated Science*
- 7 Assessment (ISA) for Oxides of Nitrogen, Oxides of Sulfur, and Particulate Matter Ecological
- 8 *Criteria*, which was then discussed at a CASAC meeting May 24-25, 2017. Comments from the
- 9 CASAC (Diez Roux, 2017) and the public were considered in preparing the second external
- 10 review draft (June 2018), which was then discussed at a CASAC meeting September 5-6, 2018
- and April 27, 2020. The CASAC provided a final letter on the second draft ISA in May 2020
- 12 (Cox, 2020), and in October 2020, the EPA released the final ISA for N oxides, SOx, and PM
- 13 ecological criteria (U.S. EPA, 2020). In August 2018, the EPA published the *Review of the*
- 14 Secondary Standards for Ecological Effects of Oxides of Nitrogen, Oxides of Sulfur, and
- 15 Particulate Matter: Risk and Exposure Assessment Planning Document (U.S. EPA 2018) which
- 16 was available for public comment (83 FR 42497, August 22, 2018).
- 17 This draft PA will be reviewed by the CASAC and available for public comment, which
- 18 will inform completion of this document and development of the Administrator's proposed
- 19 decision in this review. The current timeline projects completion of the final PA in December
- 20 2023. The timeline for the remainder of this review is governed by a consent decree that requires
- 21 the EPA to sign a notice of proposed decision by February 9, 2024, and a final decision notice by
- 22 December 10, 2024 (Center for Biological Diversity v. Regan (N.D. Cal., No. 4:22-cv-02285-
- 23 HSG)).

## 24 **1.5 ORGANIZATION OF THIS DOCUMENT**

- This PA includes staff's evaluation of the policy implications of the scientific assessment of the evidence presented and assessed in the 2020 ISA and the results of quantitative
- 27 assessments based on that information presented and assessed in this document. Taken together,
- 28 this information informs staff conclusions and the identification of policy options for
- 29 consideration in addressing public and welfare effects associated with the presence of oxides of
- 30 nitrogen, oxides of sulfur, and PM in the ambient air.
- 31 Following this introductory chapter, this document presents policy relevant information
- 32 drawn from the 2020 ISA and REA as well as assessments that translate this information into a
- 33 basis for staff conclusions as to policy options that are appropriate to consider in this review. The
- 34 discussions are generally framed by addressing policy-relevant questions that have been adapted
- 35 from those initially presented in the 2017 IRP.

1 Chapter 2 provides an overview of current information on N oxides, SOx and PM-related

2 emissions, how these pollutants are transformed in the atmosphere and contribute to deposition

3 of S and N compounds. Chapter 2 also summarizes current air concentrations and long-term

4 trends of these pollutants and associated deposition, as well as key aspects of the ambient air

5 monitoring requirements.

6 Chapter 3 reviews the basis for the existing NO<sub>2</sub> and SO<sub>2</sub> standards and outlines a general
7 approach for this review, including the additional PM secondary standard included in this
8 review.

9 In Chapter 4, we address questions related to linking ecological effects to measures that 10 can be used to characterize the extent to which such effects are reasonably considered to be 11 adverse to public welfare. This involves consideration of how to characterize adversity from a 12 public welfare perspective. In so doing, consideration is given to the concept of ecosystem 13 services, the evidence of effects on ecosystem services, and how ecosystem services can be 14 linked to ecological indicators.

15 Chapter 5 presents the exposure conditions associated with effects and the available 16 evidence providing quantitative information linking N oxides, SOx, and PM to deposition related 17 effects that can inform judgements on the likelihood of occurrence of such effects in air quality 18 conditions that meet the current standard. Quantitative analyses in this chapter help to identify 19 what effects for which the evidence is most established and robust for in regard to exposure-20 response relationships between deposition and ecosystem effects.

Chapter 6 describes the relationships between the deposition S and N compounds and air quality metrics for SOx, N oxides and PM, and other metrics with potential for effective deposition-related standards. The analyses in this Chapter are intended to characterize the relationships between ambient air concentrations and deposition particularly in rural areas, which are of most concern for this review.

26 Chapter 7 presents an assessment of the adequacy of the current NO<sub>2</sub> and SO<sub>2</sub> secondary 27 standards. Consideration is given both to the adequacy of protection afforded by the current 28 standards for both direct and deposition-related effects, as well as to the appropriateness of the 29 fundamental structure and the basic elements of the current standards for providing protection 30 from deposition-related effects. In so doing, we address questions related to considering the 31 extent to which deposition-related effects that could reasonably be judged to be adverse to public 32 welfare are occurring under current conditions which are allowed by the current standards. We 33 also consider the ways in which the structures and basic elements of the current NO<sub>2</sub> and SO<sub>2</sub> 34 secondary standards are inadequate to protect against such effects. 35 This document also includes several appendices providing additional information to

36 support the document. Appendix 5A provides an analysis conducted to compare aquatic

- 1 acidification to terrestrial acidification. Appendix 5B includes additional details related to
- 2 terrestrial ecosystem studies. This encompasses discussion of additional studies of tree growth
- 3 and survival and species richness of herb and shrub communities. Appendix 6A details the
- 4 derivation of the ecoregion air quality metrics (EAQM) for each Ecoregion/pollutant pair using
- 5 historical air quality design value (DV) data. It also describes the methodology used to calculate
- 6 the air parcel trajectories that led to the zones of influence identification, as well as the
- 7 methodologies used to estimate the EAQM values.

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37 38	Wolff, G. T. 1993. CASAC closure letter for the 1993 Criteria Document for Oxides of Nitrogen addressed to U.S. EPA Administrator Carol M. Browner dated September 30, 1993.

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 EPA Administrator Carol M. Browner dated August 22, 1995.
# **2** AIR QUALITY AND DEPOSITION

2 This chapter begins with an overview of the atmospheric processes that are relevant for 3 the review of the welfare-based secondary NAAQS for oxides of nitrogen, oxides of sulfur, 4 including those present as particulate matter (PM). This includes a description of the most 5 relevant pollutants and how they can be transformed in the atmosphere and contribute to 6 deposition of nitrogen (N) and sulfur (S) species (section 2.1). Subsequent sections summarize 7 the sources of N, S, and PM emissions (section 2.2), describe measurement of relevant species 8 including national monitoring networks and methods (section 2.3), describe recent observed 9 trends in N, S, and PM species concentrations (section 2.4), and describe the way deposition 10 estimates are developed (section 2.5).

# ATMOSPHERIC TRANSFORMATION OF NITROGEN, SULFUR, AND PM SPECIES

13 This section briefly describes the key processes associated with atmospheric deposition 14 of nitrogen and sulfur species, including both gaseous species and those that are present as PM. 15 The pathway from emission to eventual deposition is specific across pollutants and is influenced 16 by a series of atmospheric processes and chemical transformations that occur at multiple spatial 17 and temporal scales. Figure 2-1 is a simple schematic that identifies some of the individual 18 pollutants that are part of oxides of nitrogen, oxides of sulfur, and PM, and how they can be 19 interconnected. Each of these three categories of species are discussed more fully below.

1

2-1





Figure 2-1. Schematic of most relevant individual pollutants that comprise oxides of nitrogen, oxides of sulfur, and particulate matter.

### 4 2.1.1 Oxides of Sulfur

5 Sulfur dioxide (SO<sub>2</sub>) is one of a group of highly reactive gases collectively known as 6 "oxides of sulfur" (SOx). Oxides of sulfur are defined here to include sulfur monoxide (SO), 7 sulfur dioxide (SO<sub>2</sub>), sulfur trioxide (SO<sub>3</sub>), disulfur monoxide (S<sub>2</sub>O), and sulfate (in particulate 8 form as  $SO_4^{2-}$ ). As discussed in more detail in section 2.2, SOx is mostly emitted from 9 combustion processes in the form of SO<sub>2</sub>. SO<sub>2</sub> is present at higher concentrations in the ambient 10 air than the other gaseous sulfur species and as a result the NAAQS uses SO<sub>2</sub> as the indicator for 11 the larger group of SOx. Dry deposition is an important removal process for SO<sub>2</sub>. Although 12 particulate sulfate can dry deposit, it is more efficiently removed by precipitation (wet 13 deposition). 14 Once emitted to the atmosphere  $SO_2$  can react in both the gas phase and in aqueous solutions such as clouds and particles to for  $SO_4^{2-}$  (McMurry, 2004). There are multiple 15 16 pathways for this process to occur. In the daytime, atmospheric oxidation converts gas phase SO<sub>2</sub> 17 to sulfuric acid ( $H_2SO_4$ ), which quickly and nearly completely condenses on existing particles or 18 forms new sulfate particles (generically referred to as  $SO_4^{2-}$ ). The  $SO_2$  to sulfate conversion typically occurs at rates of 0.1 to 5% per hour, with higher rates associated with higher 19 20 temperatures, sunlight, and the presence of oxidants. Another important pathway is aqueous

21 phase oxidation of SO<sub>2</sub> in cloud droplets which can yield very fast rates of sulfate production.

- 1 The conversion rates are determined by the availability of oxidants. Further reactions with
- 2 ammonia form ammonium sulfate (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub>. Sulfate particles contribute to PM<sub>2.5</sub>
- 3 concentrations. The atmospheric lifetime of sulfate particles is relatively long, ranging from 2 to
- 4 10 days. As such, sulfate concentrations tend to be regionally homogeneous (see section 2.4.2).
- 5 Dry deposition is an important removal process for SO<sub>2</sub>. Although particulate sulfate can dry
- 6 deposit, it is more efficiently removed by precipitation (wet deposition).

### 7 2.1.2 Oxidized Nitrogen

- 8 The oxidized nitrogen species nitric oxide (NO) and nitrogen dioxide (NO<sub>2</sub>) are
- 9 collectively referred to as NO<sub>X</sub>. As discussed in more detail in section 2.2, the largest sources of
- 10 NO<sub>X</sub> emissions are related to fossil fuel combustion, which includes anthropogenic sources such
- 11 as power plants, industrial facilities, motor vehicles, and wood burning stoves. Non-
- 12 anthropogenic sources of NO<sub>X</sub> can include wildfires, biological soil processes, and lightning. In
- 13 the atmosphere, NO and NO<sub>2</sub> can be converted to other forms of oxidized nitrogen, including
- 14 nitric acid (HNO<sub>3</sub>), peroxynitric acid (HNO<sub>4</sub>), nitrous acid (HNO<sub>2</sub>), and peroxyacetyl nitrate
- 15 (PAN) or other forms of organic nitrogen. The term "oxides of nitrogen" refers to all forms of
- 16 oxidized nitrogen compounds (NO<sub>Y</sub>), including nitric oxide (NO), nitrogen dioxide (NO<sub>2</sub>) and all
- 17 other oxidized nitrogen-containing compounds formed from NO and NO<sub>2</sub>. The NAAQS
- 18 currently uses NO<sub>2</sub> as the indicator for the larger group of oxides of nitrogen.
- 19 There are two main pathways of nitrate formation via oxidation of NO or NO<sub>2</sub>, one which 20 occurs during the day through reaction with the hydroxyl radical to produce HNO<sub>3</sub> and the other 21 at night via reactions with other oxidants and water. Under the right thermodynamic conditions,
- some of these compounds can move from the gas phase into the solid or liquid phases as
- 23 particulate nitrate (generically referred to as NO<sub>3</sub><sup>-</sup>) and contribute to PM<sub>2.5</sub> concentrations. Each
- 24 form of oxidized nitrogen is removed from the atmosphere at different rates. For example, nitric
- 25 acid quickly settles onto surfaces (via dry deposition) while particulate nitrate is more efficiently
- 26 removed by precipitation (wet deposition).

## 27 2.1.3 Reduced Nitrogen

- 28 Distinct from oxidized nitrogen, reduced nitrogen species can contribute to PM<sub>2.5</sub>
- 29 formation and lead to adverse deposition-related effects. Ammonia (NH<sub>3</sub>) is the most common
- 30 form of atmospheric reduced nitrogen. Animal livestock operations and fertilized fields are the
- 31 largest emission sources of NH<sub>3</sub>, but there are combustion-related sources as well, such as
- 32 vehicles and fires. Ammonia plays an important role as a precursor for atmospheric particulate
- 33 matter and can be both deposited and emitted from plants and soils in a bidirectional exchange.
- 34  $NH_3$  may contribute to inorganic  $PM_{2.5}$  formation (as ammonium,  $NH_4^+$ ) based on the
- 35 availability of acid gases (HNO<sub>3</sub>, H<sub>2</sub>SO<sub>4</sub>) and favorable meteorological conditions (low

1 temperatures and high relative humidity). Ammonia reacts with gas phase nitric acid (HNO<sub>3</sub>) to

- 2 form ammonium nitrate or can partially or fully neutralize particle sulfate. The amount of
- 3 ammonia present (along with organic compounds) is one determinant of the balance of
- 4 ammonium sulfate and ammonium nitrate and therefore influences the spatial extent of N and S
- 5 deposition (ISA, Appendix 2, Section 2.3.3). Ammonia tends to dry deposit near sources, but in
- 6 particle form, ammonium  $(NH_4^+)$  can be transported farther distances and is most efficiently
- 7 removed by precipitation. The sum of  $NH_3$  and  $NH_4^+$  is referred to as  $NH_X$ .
- 8 2.1.4 Atmospheric Processing

9 Once emitted to the atmosphere, SO<sub>X</sub>, NO<sub>Y</sub>, and NH<sub>X</sub> are simultaneously impacted by 10 both chemical transformations and atmospheric transport processes until they are eventually 11 removed from the atmosphere by deposition. The transport of emitted pollutants is a function of 12 local and regional meteorological conditions such as wind fields and atmospheric stability that 13 collectively govern how the pollutant species are advected and diffused. The formation of 14 inorganic particulate matter following gas phase emission of SO<sub>X</sub>, NO<sub>Y</sub> and/or NH<sub>3</sub> is also 15 sensitive to meteorological conditions (e.g., temperature, relative humidity), and the availability 16 of basic  $(NH_3)$  or acidic  $(H_2SO_4, HNO_3)$  species. Along with the meteorological conditions, the 17 chemical lifetime of a pollutant is also a major factor in determining the distance at which 18 pollutants contribute to deposition. Since the chemical form is important to determining the rate 19 of dry and wet deposition (i.e., whether or not a pollutant deposits to plant stomata), as well as 20 the relationship between air concentrations and deposition, we use process-based models and 21 quality-assured ambient air measurements to understand the transformation from emissions to 22 concentrations to deposition (see sections 2.2 and 2.5). Additionally, landscape characteristics 23 influence deposition processes.

24

# 25 2.2 SOURCES AND EMISSIONS OF NITROGEN, SULFUR, AND PM 26 SPECIES

- 27 The sources and precursors to gaseous and particulate forms of SO<sub>X</sub>, NO<sub>Y</sub>, and NH<sub>X</sub> vary
- and can originate from a combination of manmade and natural sources. Anthropogenic sources
- of air pollutants that result in adverse deposition-related effects (i.e., SO<sub>2</sub>, NO<sub>X</sub>, and NH<sub>3</sub>)
- 30 include power plants, industrial sources, motor vehicles, and agriculture. The National Emissions
- 31 Inventory (NEI)<sup>1</sup> is a comprehensive and detailed estimate of air emissions of criteria pollutants,
- 32 precursors to criteria pollutants, and certain hazardous air pollutants from air emissions sources.

<sup>&</sup>lt;sup>1</sup> <u>https://www.epa.gov/air-emissions-inventories/national-emissions-inventory-nei</u>

1 The NEI is released every three years based primarily upon data provided by State, Local, and

- 2 Tribal air agencies for sources in their jurisdictions and supplemented by data developed by the
- 3 EPA. For some sources, such as power plants, direct emission measurements enable the
- 4 emissions estimates to be more certain than other sectors without such direct measurements. It
- 5 should be recognized that emission inventories contain assumptions that may influence the
- 6 estimates of their magnitude and trends. The 2020 NEI was released to the public on March 31,
- 7 2023. These 2020 data will be used for the summaries shown in the following sections describing
- emission estimates and trends. The reader is referred to the 2020 NEI<sup>2</sup> for further details. 8
- 9

# 2.2.1 NOx Emissions Estimates and Trends

10 Figure 2-2 shows the relative contributions of various sources to total U.S.  $NO_X$ 11 emissions in 2020, based on estimates contained in the EPA NEI (2023). Anthropogenic sources 12 account for a majority of NO<sub>X</sub> emissions in the U.S., with highway vehicles (26%), stationary 13 fuel combustion (25%), and non-road mobile sources (19%) identified as the largest contributors 14 to total emissions. Highway vehicles include all on-road vehicles, including light duty as well as 15 heavy duty vehicles, both gasoline- and diesel-powered. The stationary fuel combustion sector 16 includes electricity generating units (EGUs), as well as commercial, institutional, industrial, and 17 residential combustion of biomass, coal, natural gas, oil, and other fuels. Non-road mobile 18 sources include aircraft, commercial marine vessels, locomotives, and non-road equipment. 19 Other anthropogenic NO<sub>X</sub> sources include agricultural field burning, prescribed fires, and various 20 industrial processes such as cement manufacturing and oil and gas production. Natural sources of 21 NO<sub>X</sub> include emissions from plants and soil (biogenic) which represent 12% of the total NO<sub>X</sub> 22 emissions. In sum, fires (i.e., wild, prescribed, and agricultural) are estimated to represent 5% of 23 the overall emissions of NO<sub>X</sub>. 24 Figure 2-3 shows the NO<sub>X</sub> emissions density in tons/year per square mile for each U.S. 25 County. The majority of NO<sub>X</sub> emissions tend to be located near urban areas, which tend to have

26 the most vehicle traffic and industrial sources. However, there are also some counties in rural

27 areas with higher NO<sub>x</sub> emissions due to the presence of large stationary sources such as EGUs or

28 oil and gas extraction and generation.

<sup>&</sup>lt;sup>2</sup> https://www.epa.gov/air-emissions-inventories/2020-national-emissions-inventory-nei-data





Figure 2-2. 2020 NOx emissions estimates by source sector (U.S. EPA NEI, 2023).





- 1 Total NO<sub>X</sub> emissions have trended strongly downward across the U.S. between 2002 and
- 2 2022. Nationwide estimates indicate a 70% decrease in anthropogenic  $NO_X$  emissions over this
- 3 time period as a result of multiple regulatory programs (e.g., including the NO<sub>X</sub> SIP Call, the
- 4 Cross-State Air Pollution Rule (CSAPR), and the Tier 3 Light-duty Vehicle Emissions and Fuel
- 5 Standards) implemented over the past two decades, as well as changes in economic conditions.
- 6 As seen in Figure 2-4, the overall decrease in NO<sub>X</sub> emissions has been driven primarily by
- 7 decreases from the three largest emissions sectors. Specifically, compared to the 2002 start year,
- 8 estimates for 2022 (from the 2020 NEI) indicate an 84% reduction in NO<sub>X</sub> emissions from
- 9 highway vehicles, a 68% reduction in NO<sub>X</sub> emissions from stationary fuel combustion, and a
- 10 54% reduction in NO<sub>X</sub> emissions from non-road mobile sources.
- 11



- 12
- 13 Figure 2-4. Trends in NOx emissions by sector between 2002 and 2022.

14

### 1 2.2.2 SO<sub>2</sub> Emissions Estimates and Trends

2 Fossil fuel combustion is the main anthropogenic source of SO<sub>2</sub>, primarily from coal-3 fired EGUs (48%). Sulfur is present to some degree in all fossil fuels, especially coal, and occurs 4 as reduced organosulfur compounds. Of the most common types of coal (anthracite, bituminous, 5 subbituminous, and lignite), sulfur content varies between 0.4 and 4% by mass. Sulfur in fossil 6 fuels is almost entirely converted to SO<sub>2</sub> during combustion. Other major anthropogenic sources 7 of SO<sub>2</sub> emissions include industrial processes (27%) and stationary source fuel combustion (9%). 8 Mobile sources, and agricultural and prescribed fires are smaller contributors. Figure 2-5 shows 9 the percentage contribution of specific source categories to the total anthropogenic (plus wildfire) SO<sub>2</sub>. 10

- 11 Figure 2-6 shows the SO<sub>2</sub> emissions density in tons/year per square mile for each U.S.
- 12 county. The majority of SO<sub>2</sub> emissions tend to be located near large point sources such as coal-
- 13 fired EGUs or large industrial facilities. Counties near urban areas also tend to have higher SO<sub>2</sub>
- 14 emissions due to the higher concentration of industrial facilities. In some cases, counties in rural
- 15 areas can also have higher emissions due to oil and gas extraction or fires.



May 2023



# 2 Figure 2-6. 2020 SO<sub>2</sub> emissions density across the U.S. (U.S. EPA NEI, 2023).

3 Similar to NO<sub>X</sub>, and for many of the same reasons, SO<sub>2</sub> emissions have declined

4 significantly since 2002. Figure 2-7 illustrates the emissions changes over the 2002-2022 period.

5 The data shows an 87% decrease in total SO<sub>2</sub> emissions over the period, including reductions of

6 91% in emissions from EGUs and 96% in emissions from mobile sources.

7





### 3 2.2.3 NH<sub>3</sub> Emissions Estimates and Trends

4 NH<sub>3</sub> is directly emitted, differing from other atmospheric N species (e.g., organic N, 5 NO<sub>2</sub>) that are formed through photochemical reactions. Figure 2-8 shows the percentage 6 contribution of specific source categories to the total anthropogenic (plus wildfires) NH<sub>3</sub>. In 7 2020, livestock waste (49%), fertilizer application (33%) and aggregate fires (11%) contributed 8 most significantly to total annual emissions (5.5 million tons NH<sub>3</sub>). While mobile source 9 contributions to total NH<sub>3</sub> emissions are only about 2% at the national level, there is a growing 10 body of evidence suggesting that vehicular sources may be underestimated in the NEI (Sun et al., 11 2017; Chen et al., 2022). Any underestimation in mobile source NH<sub>3</sub> emissions would mostly 12 impact urban areas, where there is a lot of on-road mobile source traffic. Figure 2-9 shows the 13 NH<sub>3</sub> emissions density in tons/year per square mile for each U.S. county. Ammonia emissions 14 are greatest in counties with significant agricultural output (e.g., central U.S., parts of CA, and 15 eastern NC.



1 2

Figure 2-8. 2020 NH<sub>3</sub> emissions by source sector (U.S. EPA NEI, 2023).





Figure 2-9. NH<sub>3</sub> Emissions density across the U.S. (U.S. EPA NEI, 2023).

1 Figure 2-10 shows NH<sub>3</sub> emission trends from 2002-2022. In comparison with NOx and 2 SOx emission trends, which demonstrated dramatic decreases over the past few decades, the annual rate of NH3 emissions remained relatively flat with even a noted upward trend in recent 3 4 years. However, there is greater uncertainty in NH<sub>3</sub> emissions trends (ISA, Appendix 2, section 5 2.2.3). This is partly due to a lack of control programs nationally for agricultural sources of NH<sub>3</sub>. 6 It is worth noting that variabilities associated with local management practices related to animal 7 husbandry makes these emissions a bit more uncertain than emissions, for example, derived from 8 a mobile source model or direct measurements from EGU sources. The EPA has built improved 9 models for both livestock waste emissions and fertilizer application process to inform the 2020 10 NEI which is expected to have reduced these uncertainties. The reader is referred to our 2020 NEI Technical Support Document (TSD) (U.S. EPA, 2023). 11





# 2.3 MONITORING AMBIENT AIR CONCENTRATIONS AND DEPOSITION OF N, S, AND PM

To promote uniform enforcement of the air quality standards set forth under the CAA, the EPA has established federal reference methods (FRMs) and federal equivalent methods (FEMs) for ambient air sample collection and analysis. Measurements for determinations of NAAQS compliance must be made with FRMs or FEMs. FRMs have been established and national monitoring networks put in place for NO<sub>2</sub> as the indicator of oxides of nitrogen, SO<sub>2</sub> as the indicator of sulfur oxides, and PM<sub>2.5</sub> and PM<sub>10</sub> as indicators for PM.

9 As described briefly below, multiple monitoring networks measure the atmospheric 10 concentrations of nitrogen oxides, SO<sub>x</sub>, and PM, as well as wet deposition of N and S. The 11 largest routinely operating network measuring ambient air concentrations is the State and Local 12 Air Monitoring Stations (SLAMS) network which includes measurement of one or more 13 NAAQS pollutants at each site. There are three multipollutant networks involving NAAQS 14 measurements which are largely sited at SLAMS<sup>3</sup>. These networks include: the National Core 15 (NCore) multi-pollutant monitoring network, the Photochemical Assessment Monitoring Stations 16 (PAMS) network, and the Near-Road network. The NCore network is notable in that it provides 17 a core of sites, mostly located in urban areas, that provide co-located measurements of SO<sub>2</sub>, NO, 18 NO<sub>Y</sub>, and PM components including ammonium, nitrate, and sulfate, although with sparser 19 coverage than the FRM networks for SO<sub>2</sub> or NO<sub>2</sub>. Because NO<sub>Y</sub> is measured rather than NO<sub>X</sub>, and because of collocated SO<sub>2</sub> and SO<sub>4</sub><sup>2-</sup> measurements, ambient air concentrations of both NO<sub>Y</sub> 20 21 and SO<sub>X</sub> can be determined from NCore data, so that these data can be used to help estimate total 22 deposition of oxides of nitrogen and sulfur. The primary objective of the PAMS network is to 23 support the implementation of the ozone NAAQS, it also measures NO<sub>Y</sub> as well as having a 24 requirement to measure NO<sub>2</sub>. The Near-road network is intended to capture short-term peak NO<sub>2</sub> 25 concentrations for comparison to the NAAQS. Many of the Near-Road sites are also required to 26 have collocation with PM<sub>2.5</sub> and carbon monoxide (CO). One of the challenges associated with 27 interpreting monitoring data in the context of a deposition-related secondary standard is that 28 many, but not all, of the monitor sites are located in urban or suburban areas, while many of the 29 areas where adverse deposition effects are of greatest concern tend to be in more rural areas.

30

2.3.1 NOx Monitoring Networks

31 There were 491 monitoring sites reporting hourly NO<sub>2</sub> concentration data to the EPA

32 during the 2019-2021 period; 80% of these NO<sub>2</sub> monitoring sites are part of the SLAMS

33 network. This network relies on a chemiluminescent FRM and on multiple FEMs that use either

<sup>&</sup>lt;sup>3</sup> A small number of multipollutant sites may have a monitor type different than SLAMS such as Tribal or Non-EPA Federal (e.g., National Park Service [NPS]).

- 1 chemiluminescence or direct measurement methods of NO<sub>2</sub>. Chemiluminescent-based FRMs
- 2 only detect NO in the sample stream. Therefore, a two-step process is employed to measure NO<sub>2</sub>,
- 3 based on the subtraction of NO from NO<sub>X</sub>. Data produced by chemiluminescent analyzers
- 4 include NO, NO<sub>2</sub>, and NO<sub>X</sub> measurements. As discussed in the ISA (U.S. EPA, 2020, p. 2-34)
- 5 the traditional chemiluminescence FRM is subject to potential measurement biases resulting
- 6 from interference by N oxides other than NO or NO<sub>2</sub>.<sup>4</sup> These potential biases are measurement
- 7 uncertainties that can impact exposure analyses. However, within metropolitan areas, where a
- 8 majority of the NO<sub>2</sub> monitoring network is located and is influenced by strong NO<sub>X</sub> sources, the
- 9 potential for bias related to other N oxides is relatively small.
- 10 Another important subset of SLAMS sites is the near-road monitoring network, which 11 was required as part of the 2010 NO<sub>2</sub> NAAQS review and began operating in 2014. Near-road
- 12 sites are required in each metropolitan statistical area (MSA) with a population of 1,000,000 or
- 12 sites are required in each metropolitan statistical area (MSA) with a population of 1,000,000 of
- 13 greater, and an additional near-road site is required in each MSA with a population of 2,500,000
- 14 or greater. There were 73 near-road monitors in operation during the 2019-2021 period. Finally,
- 15 there are also a number of Special Purpose Monitors (SPMs), which are not required but are
- 16 often operated by air agencies for short periods of time (i.e., less than 3 years) to collect data for
- 17 human health and welfare studies, as well as other types of monitoring sites, including monitors
- 18 operated by tribes and industrial sources. The SPMs are typically not used to assess compliance
- 19 with the NAAQS. The locations of all NO<sub>2</sub> monitoring sites operating during the 2019-2021
- 20 period are shown in Figure 2-11.
- 21

<sup>&</sup>lt;sup>4</sup> The N oxides other than NO and NO<sub>2</sub> are often collectively abbreviated as NO<sub>2</sub> (i.e., NO<sub>Y</sub> = NO<sub>X</sub> +NO<sub>Z</sub>).



SLAMS (250)

● NCORE/PAMS (74) ● NEAR ROAD (73) ● SPM/OTHER (94)



3

### 2.3.2 SO<sub>2</sub> Monitoring Networks

4 There were 505 monitoring sites reporting hourly SO<sub>2</sub> concentration data to the EPA 5 during the 2019-2021 period. Over 75% of the SO<sub>2</sub> sites are part of the SLAMS network. 6 Measurements are made using ultraviolet fluorescence (UVF) instruments, which are designated 7 as FRMs or FEMs and the data are reported as hourly concentrations with either the maximum 5-8 minute concentration for each hour or twelve 5-minute average concentrations for each hour. 9 Additionally, as of 2015, States are required to monitor or model ambient air  $SO_2$  levels in areas 10 with stationary sources of SO<sub>2</sub> emissions of over 2,000 tons per year. The EPA identified over 11 300 sources meeting these criteria according to 2014 emissions data, and some States chose to 12 set up ambient air monitoring sites to assess compliance with the SO<sub>2</sub> NAAQS. Some of these 13 monitors are operated by the States as SLAMS monitors, while others are operated by the 14 industrial sources. The locations of all SO<sub>2</sub> monitoring sites (FRM or FEM) operating during the 15 2019-2021 period are shown in Figure 2-12.





## 3 2.3.3 PM<sub>2.5</sub> Monitoring Networks

4 As with NOx and SO<sub>2</sub>, the main network of monitors providing ambient air PM mass data for 5 use in NAAOS implementation activities is the SLAMS network (including NCore). PM<sub>2.5</sub> 6 monitoring was required for near-road network sites as part of the 2012 PM NAAQS review and 7 these sites monitors were phased into the network between 2015 and 2017. Near-road sites are 8 also required in each MSA with a population of 1,000,000 or greater. The PM<sub>2.5</sub> monitoring 9 program remains one of the largest ambient air monitoring programs in the U.S. There were 1,067 monitoring sites reporting PM<sub>2.5</sub> data to the EPA during the 2019-2021 period. Figure 2-13 10 11 shows the locations of these monitoring sites. Approximately 50% of these monitoring sites operate automated FEMs which report continuous (hourly) PM<sub>2.5</sub> data while the remaining sites 12 operate FRMs which collect 24-hour samples every day, every 3<sup>rd</sup> day, or every 6<sup>th</sup> day. 13 14





3 Due to the complex nature of fine particles, the EPA and States implemented the 4 Chemical Speciation Network (CSN) to better understand the components of fine particle mass 5 at selected locations across the country. PM<sub>2.5</sub> speciation measurements are also collected at 6 NCore stations. Additionally, specific components of fine particles are measured through the 7 Interagency Monitoring of Protected Visual Environments (IMPROVE) monitoring program, 8 which supports the regional haze program and tracks changes in visibility in Federal Class I 9 areas as well as many other rural and some urban areas. The IMPROVE network consists of 10 more than 100 monitoring sites in national parks and other remote locations and has also 11 provided a reliable, long-term record of particulate mass and species components. The locations 12 of the CSN (3-day frequency) and IMPROVE (6-day frequency) sites reporting speciated PM<sub>2.5</sub> 13 data to the EPA during the 2019-2021 period are shown in Figure 2-14. 14



CSN (107)
 IMPROVE (151)
 NCORE (49)
 OTHER (9)



#### 3 2.3.4 Other Monitoring Networks Relevant to N, S, and PM Deposition

4 Wet deposition is measured as the product of pollutant concentration in precipitation and 5 precipitation amounts (e.g., in rain or snow). Concentration in precipitation is currently measured as a weekly average by the National Atmospheric Deposition Program/National Trends Network 6 7 (NADP/NTN) across a national network of approximately 250 sites using a standard 8 precipitation collector. The NADP precipitation network was initiated in 1978 to collect data on 9 amounts, trends, and distributions of acids, nutrients, and cations in precipitation. The NTN is 10 the only network (shown in Figure 2-15) that provides a long-term record of precipitation chemistry across the U.S. Sites are mainly located away from urban areas and pollution sources. 11 12 An automated collector ensures that the sample is exposed only during precipitation (wet-only 13 sampling). Nitrate, sulfate, and ammonium are all measured. Relatively high confidence has been 14 assigned to wet deposition estimates because of established capabilities for measuring relevant 15 chemical components in precipitation samples.

16



Figure 2-15. Location of NTN monitoring sites with sites active shown in blue and inactive sites in white.

1 In contrast, direct measurements of dry deposition flux are rare and difficult, and dry

- 2 deposition fluxes of gases and particles are estimated from concentration measurements by an
- 3 inferential technique described in the 2008 ISA (U.S. EPA, 2008). Ambient air concentrations
- 4 are measured in the Clean Air Status and Trends Network (CASTNET), which was established
- 5 under the 1991 Clean Air Act Amendments to assess trends in acidic deposition. CASTNET is a
- 6 long-term environmental monitoring network with approximately 100 sites (see Figure 2-16 for a
- 7 map of U.S. sites) located throughout the U.S. and Canada, managed and operated by the U.S.
- 8 EPA in cooperation with other federal, state, and local partners (<u>www.epa.gov/castnet</u>).



9 10

11 12

**Figure 2-16.** Location of CASTNET monitoring sites and the organizations responsible for collecting data. (NPS = National Park Service, BLM = Bureau of Land Management)

CASTNET is the only network in the U.S. that provides a consistent, long-term data record of ambient air concentrations of S and N species that dry deposition fluxes can be estimated from. It complements the NTN, and nearly all CASTNET sites are collocated with or near an NTN site. Together, these two monitoring programs are designed to provide data

17 necessary to estimate long-term temporal and spatial trends in total deposition (dry and wet).

1 Species measured in CASTNET include: O<sub>3</sub>, SO<sub>2</sub>, HNO<sub>3</sub>, nitrate, sulfate, and ammonium among

- 2 others. Weekly ambient air concentrations of gases and particles are collected with an open-face
- 3 3-stage filter pack. Ozone measurements occur on an hourly basis. While CASTNET data are
- 4 more useful for estimating dry deposition than data from FRM networks, monitors are generally
- 5 sparse and deposition is only determined for discrete locations. Also, not all of the species that
- 6 contribute to total sulfur and nitrogen deposition are measured in CASTNET (Schwede et al.,
- 7 2011). Despite these disadvantages, CASTNET data still be very useful if used in combination
- 8 with modeled data (Schwede et al., 2011) as discussed further in Section 2.5.

9 There are differences in the measurement techniques that require careful consideration 10 when used for analysis. IMPROVE and CSN are most efficient at collecting particles with a 11 diameter smaller than 2.5 microns (PM<sub>2.5</sub>), while the CASTNET samplers, which do not use 12 size-selected inlets, also measure larger particles. This is relevant because larger particles are 13 often from wind-blown soil, dust, or sea salt. Gas-phase nitric acid can condense onto these 14 particles, forming particulate nitrate. Since these larger particles deposit quickly, this can be a 15 significant portion of the total N deposition. However, as most CASTNET sites are located in 16 rural areas, the expectation is that unless these sites are disproportionately impacted by local 17 coarse particle sources, that most of the PM collected is PM<sub>2.5</sub>. Furthermore, the timing of the 18 measurements is not the same. CASTNET filter packs are deployed in the field for the entire 7-19 day measurement period, while IMPROVE and CSN are 24-hour measurements. Since 20 ammonium nitrate is semi-volatile, and as temperature and humidity conditions change, these 21 particles can evaporate off the filter as gas-phase ammonia and nitric acid. Each network deploys 22 a different approach to minimizing these evaporative losses or capturing the volatilized nitrate 23 and ammonia (Lavery et al., 2009). When co-located and compared to reference techniques, the 24 correlation between these measurement techniques depends on meteorological conditions. Due to 25 large measurement artifacts, IMPROVE no longer reports ammonium (NH4<sup>+</sup>), and CASTNET 26 reports the sum of nitric acid and particle nitrate (total NO<sub>3</sub>) as a more certain measurement. 27 The NADP also maintains the Ammonia Monitoring Network (AMoN) which is designed 28 to capture long-term trends in ambient air NH<sub>3</sub> concentrations and deposition. There are 29 currently 106 AMoN sites covering 34 states (see Figure 2-17). The AMoN uses passive filter-30 based samplers which are deployed for two-week periods. Both gaseous ammonia and particle 31 ammonium concentrations are measured. 32





Figure 2-17. Location of AMoN monitoring sites with sites active shown in blue and inactive sites in white. (There is an additional site in AK not shown here.)

## 1 2.4 RECENT AMBIENT AIR CONCENTRATIONS AND TRENDS

#### 2 2.4.1 NO<sub>2</sub> Concentrations and Trends

3 There are currently two forms of the primary NO<sub>2</sub> NAAQS. One is based on the 98<sup>th</sup> 4 percentile of the 1-hour daily maximum concentrations averaged over 3 years and the level is set 5 at 100 ppb. The other is based on the annual mean and the level of the standard is set at 53 ppb. 6 The secondary NO<sub>2</sub> NAAQS is also based on the annual mean with the same level of 53 ppb. As 7 shown in Figures 2-18 and 2-19, there are no locations with NO<sub>2</sub> design values in violation of 8 these standards. The highest NO<sub>2</sub> concentrations mostly occur in urban areas across the western 9 U.S. (e.g., Los Angeles, Phoenix, Las Vegas, Denver). The maximum 1-hour design value during 10 the 2019-2021 period was 80 ppb, while the annual design value for 2021 was 30 ppb. Both 11 maximum design values occurred at near-road sites in the Los Angeles metropolitan area. For the 12 2019-2021 period, the mean average hourly NO<sub>2</sub> value, across valid monitoring sites, was 16.3 13 ppb.

14 NO<sub>2</sub> concentrations have been declining across the U.S. for decades, in response to 15 cleaner motor vehicles, emissions reductions at stationary fuel combustion sources, and 16 economic factors. For example, in Los Angeles metropolitan area annual NO2 design values 17 were almost twice as high in the early 1980's (U.S. EPA, 1985). Figures 2-20 and 2-21 show the 18 trends in the annual 98<sup>th</sup> percentile of the daily maximum 1-hour NO<sub>2</sub> concentrations and in the 19 annual mean NO<sub>2</sub> concentrations across the U.S. going back to 1980. The trends are sharply 20 downward for both forms of the NO<sub>2</sub> standard. At the beginning of the trends record, it was not 21 uncommon for locations to exceed the NO<sub>2</sub> NAAQS, especially the standard with the shorter 22 averaging time. However, the last violations of the NO<sub>2</sub> standards occurred in 1991 (annual) and 23 2008 (hourly). Over the past decade, the downward trends in  $NO_2$  levels across the U.S. have 24 continued, but at a slower rate than what was experienced from 1980 to 2010. Given that 25 deposition-related impacts can adversely affect ecosystems (forests/trees, streams/fish) over the 26 course of decades (as discussed in more detail in Section 5 of this assessment), it is important to 27 recognize that effects of the high NO<sub>2</sub> levels observed in 1980, and preceding decades when NO<sub>2</sub> 28 levels were even higher, may still be impacting ecosystem health. Prior to 1980, the monitoring 29 networks were somewhat sparser, but NO<sub>2</sub> data exist for certain cities. The EPA's very first 30 Trends Report (U.S. EPA, 1973) reported annual average NO<sub>2</sub> values in five U.S. cities for the 31 1967-1971 period. At that time, annual average NO<sub>2</sub> concentrations averaged 75 ppb over the 32 cities where data existed (i.e., off the chart of the 1980-2021 trend shown in Figure 2-21). See 33 Table 2-1 for a summary of these older NO<sub>2</sub> annual means.



Figure 2-18. Primary NO<sub>2</sub> design values (98<sup>th</sup> percentile of daily maximum 1-hourly concentrations, averaged over 3 years; ppb) at monitoring sites with valid design values for the 2019-2021 period.



2 3

• 1 - 10 ppb (297 sites) • 11 - 20 ppb (99 sites) • 21 - 30 ppb (8 sites)





Figure 2-20. Distributions of annual 98th percentile, maximum 1-hour NO<sub>2</sub> design values (ppb) at U.S. sites across the 1980-2021 period. The red line shows the number of sites included in each boxplot per year.



Figure 2-21. Distributions of annual mean NO<sub>2</sub> design values (ppb) at U.S. sites across the 1980-2021 period. The red line shows the number of sites included in each boxplot per year.

May 2023

Location	1967-1971 Annual Mean NO₂ Concentration (ppb)
Chicago	120.5
Cincinnati	60.4
Denver	65.1
Philadelphia	76.1
St. Louis	54.1
5-city average	75.3

1Table 2-1.Average annual mean NO2 concentrations in select cities for the 1967-19712period.

#### 3 2.4.2 SO<sub>2</sub> Concentrations and Trends

The primary SO<sub>2</sub> standard is based on the 99<sup>th</sup> percentile of daily maximum 1-hour 4 5 concentrations, averaged over 3 years, and is currently set at a level of 75 ppb. The secondary 6 SO<sub>2</sub> standard uses an averaging time of 3 hours with a level of 0.5 ppm (500 ppb) and the form 7 of the standard is that the level is not to be exceeded more than once per year. As shown in 8 Figure 2-22, for the 2019-2021 period, there were 15 locations with SO<sub>2</sub> design values in 9 violation of the primary SO<sub>2</sub> standard. The maximum design value was 376 ppb at a monitoring 10 site near an industrial park in southeast Missouri. The sites with design values exceeding the 11 NAAQS in Hawaii are due to natural SO<sub>2</sub> emissions from recurring volcanic eruptions. Both 12 peak and mean SO<sub>2</sub> concentrations are higher at source-oriented monitoring sites than non-13 source sites. Mean hourly SO<sub>2</sub> concentrations are 3 ppb (5.1 ppb at source-oriented sites, 1.6 ppb 14 at urban non-source sites, and 0.9 ppb at rural non-source sites). Figure 2-23 displays the second 15 highest 3-hourly SO<sub>2</sub> values across the U.S. in 2021. All sites with valid secondary SO<sub>2</sub> design 16 values were less than the 500 ppb level and the vast majority of sites had concentrations that 17 were less than 20 ppb. Like NO<sub>2</sub>, SO<sub>2</sub> concentrations have been declining across the U.S. for 18 decades, primarily in response to emissions reductions at stationary fuel combustion sources. 19 Figure 2-24 shows the downward trend in design values for the primary SO<sub>2</sub> NAAQS over the 20 past 40 years. 1994 was the last year in which the median site had a design value greater than the 21 current primary 1-hour standard of 75 ppb. Since then, the entire distribution of values has 22 continued to decline such that the median values across the network of sites is now less than 10 23 ppb. Additional sites were added to the network in 2017 near major industrial sources of SO<sub>2</sub> and 24 this likely caused the slight increase in the median concentration observed in 2017. Finally, 25 Figure 2-25 shows the sharp downward trend in annual SO<sub>2</sub> concentrations across the U.S. 26 Again, the highest values in the distribution in recent years are from the sites near industrial 27 sources.

2-26



Figure 2-22. Primary SO<sub>2</sub> standard design values (99<sup>th</sup> percentile of 1-hour daily maximum concentrations, averaged over 3 years; ppb) for the 2019-2021 period at monitoring sites with valid design values.





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Figure 2-23. Secondary SO<sub>2</sub> standard design values (2<sup>nd</sup> highest 3-hourly average; ppb)
 for the year 2021 at monitoring sites with valid design values.



**Figure 2-24.** Distributions of 99th percentile of maximum daily 1-hour SO<sub>2</sub> design values (ppb) at U.S. sites across the 1980-2021 period. The red line shows the number of sites included in each boxplot per year. Orange dots represent design values in Hawaii determined to have been influenced by volcanic emissions. Note: the y-axis is plotted on a logarithmic scale.



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Figure 2-25. Distributions of annual average SO<sub>2</sub> design values (ppb) at U.S. sites across
 the 2000-2021 period. Sites from Hawaii are not included.

### 1 2.4.3 PM<sub>2.5</sub> Concentrations and Trends

- 2 There are three relevant standards for  $PM_{2.5}$ . There are two standards based on annual
- 3 means, averaged over 3 years, with levels at 12.0  $\mu$ g/m<sup>3</sup> (primary standard) and 15.0  $\mu$ g/m<sup>3</sup>
- 4 (secondary standard). There is also a 24-hour standard (both primary and secondary) that is
- 5 based on the 98<sup>th</sup> percentile of daily PM<sub>2.5</sub> values, averaged over 3 years, with a level of 150
- $6 \mu g/m^3$  that is not to be exceeded more than once per year. As discussed in Section 2.1, PM<sub>2.5</sub> is a
- 7 mixture of substances suspended as small liquid and/or solid particles. Figure 2-26 displays a
- 8 map with pie charts showing the major  $PM_{2.5}$  species as a fraction of total  $PM_{2.5}$  mass as
- 9 measured at selected NCore, CSN, and IMPROVE sites during the 2019 to 2021 period. The six
- 10 species shown are sulfate  $(SO_4^{2-})$ , nitrate  $(NO_3^{-})$ , elemental carbon (EC), organic carbon (OC),
- 11 crustal material, and sea salt. The mix of PM<sub>2.5</sub> components can vary across the U.S. For
- 12 example, in the Appalachian region, the predominant contributor to total PM<sub>2.5</sub> mass is sulfate.
- 13 Conversely, in the upper Midwest, the largest component term tends to by nitrate. This regional
- 14 variability in PM<sub>2.5</sub> composition has implications for the spatial nature of N and S deposition.



Figure 2-26. Map showing pie charts of PM<sub>2.5</sub> component species at selected U.S.
 monitoring sites based on 2019-2021 data.

- 18 Figures 2-27 and 2-28 show maps of the annual and 24-hour PM<sub>2.5</sub> design values,
- 19 respectively, at U.S. ambient air monitoring sites based on monitoring data from the 2019-2021
- 20 period. All sites in the eastern U.S. were meeting both the annual primary PM<sub>2.5</sub> NAAQS of 12.0
- 21  $\mu g/m^3$  and the 24-hour PM<sub>2.5</sub> NAAQS of 35  $\mu g/m^3$  during this period. Many sites in the western
- 22 U.S. were still violating the 24-hour PM<sub>2.5</sub> NAAQS in 2019-2021, while a smaller number of

1 sites, mostly in California, were also violating the annual  $PM_{2.5}$  NAAQS (28 sites which exceed 2 the primary NAAQS level of 12.0  $\mu$ g/m<sup>3</sup>, and 9 sites which exceed the secondary annual PM<sub>2.5</sub> 3 NAAQS level of 15.0  $\mu$ g/m<sup>3</sup>). It should be noted that large areas of the western U.S. were 4 impacted by smoke from wildfires in 2020 and 2021 and these smoke-impacted concentrations 5 are included in the 2019-2021 data shown here. The highest annual PM<sub>2.5</sub> design values are 6 located in the San Joaquin Valley of California, while the highest 24-hour PM2.5 design values 7 are located in Mono County, California, which was heavily impacted by wildfire smoke in 2020. 8 Figures 2-29 and 2-30 display the average nitrate and sulfate concentrations over the U.S. 9 during the period 2019-2021. As discussed above, sulfate concentrations are highest in the Ohio 10 River valley and along the Gulf of Mexico, while nitrate concentrations are highest in the upper 11 Midwest, along the northeast urban corridor, and in parts of California. Figures 2-31 and 2-32 12 show trends in annual average concentrations for nitrate and sulfate based on sites that collected 13 data for at least 12 out of 16 years from 2006 to 2021. Broad national reductions in  $NO_X$ 14 emissions have resulted in significant decreasing trends in nitrate concentrations in most of the 15 U.S., especially in areas where nitrate concentrations were historically highest. Similarly, 16 reductions in SO<sub>2</sub> emissions have resulted in significant reductions in sulfate concentrations 17 nationally and especially in the eastern U.S. National, annual average  $PM_{2.5}$  concentrations have 18 declined despite the relatively consistent trend in NH<sub>3</sub> emissions. While not shown here, trends 19 in other PM<sub>2.5</sub> components like EC and OC were more variable, with some sites showing 20 significant decreases and the remaining sites having no clear trend. Ammonium sulfate and 21 ammonium nitrate make up less than one-third of the  $PM_{2.5}$  mass at the majority of sites and only 22 a few sites have more than half of the PM<sub>2.5</sub> mass from these compounds. 23 The EPA has also promulgated standards for  $PM_{10}$  (a 24-hour primary and secondary 24 standard with a level of 150  $\mu$ g/m<sup>3</sup> that is not to be exceeded more than once per year, averaged 25 over three years). While PM<sub>2.5</sub> mass is composed mainly of sulfates, nitrates, and other organic 26 matter that can contribute to ecosystem impacts (ISA, Appendix 2, Section 2.1), PM<sub>10-2.5</sub> is 27 mostly composed of crustal material as well as sea salt in coastal areas. There is little discussion

- 28 of PM<sub>10-2.5</sub> effects in this document because these particles have faster settling velocities and the
- 29 composition of this mass is expected to have less impact on deposition-related welfare impacts.



Figure 2-27. Primary and secondary annual PM<sub>2.5</sub> design values (annual mean, averaged over 3 years, 2019-2021 period) at monitoring sites with valid design values.



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5 Figure 2-28. Primary and secondary 24-hour PM<sub>2.5</sub> design values (98<sup>th</sup> percentile, averaged 6 over 3 years; 2019-2021 period) at monitoring sites with valid design values.



2 Figure 2-29. Average NO<sub>3</sub><sup>-</sup> concentrations (µg/m<sup>3</sup>) for the 2019-2021 period.



4 Figure 2-30. Average  $SO_4^{2-}$  concentrations ( $\mu g/m^3$ ) for the 2019-2021 period.



Decreasing > 0.1 ug/m^3/yr (11 sites)
 No Significant Trend (102 sites)
 Decreasing < 0.1 ug/m^3/yr (138 sites)</li>

Figure 2-31. Trends in annual average concentrations for nitrate (NO<sub>3</sub><sup>-</sup>) from 2006 through 2021.



4

′ Decreasing > 0.1 ug/m^3/yr (108 sites) ▼ Decreasing < 0.1 ug/m^3/yr (146 sites)

5 Figure 2-32. Trends in annual average concentrations for sulfate (SO<sub>4</sub><sup>2-</sup>) from 2006 through 2021.

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1 The trends in total  $PM_{2.5}$  mass between 2000 and 2021 are shown in Figures 2-33 (annual 2 standard) and 2-34 (24-hour standard). These plots show the national distribution of PM<sub>2.5</sub> concentrations, along with the number of PM<sub>2.5</sub> monitoring sites reporting data in each year. The 3 4 median of the annual average PM<sub>2.5</sub> concentrations decreased by 38 percent, from 12.8  $\mu$ g/m<sup>3</sup> in 2000 to 8  $\mu$ g/m<sup>3</sup> in 2021. Similarly, the median of the annual 98<sup>th</sup> percentile 24-hour PM<sub>2.5</sub> 5 concentrations decreased by 35 percent, from 32  $\mu$ g/m<sup>3</sup> in 2000 to 21  $\mu$ g/m<sup>3</sup> in 2021. Both the 6 annual average and 98<sup>th</sup> percentile 24-hour PM<sub>2.5</sub> concentrations decreased steadily from the 7 8 early 2000s until 2016, and have fluctuated in recent years, especially in the upper tail of the 9 distribution. These fluctuations are largely due to large-scale wildfire events that have occurred 10 in recent years. The size of the PM2.5 monitoring network increased rapidly following the 11 establishment of a PM<sub>2.5</sub> NAAQS in 1997, and the network has been relatively stable at around 12 1,200 sites since 2002.

# 13 2.4.4 Ammonia Concentrations and Trends

14 The AMoN network has collected measurements of ammonia gas since 2010 (NADP, 15 2011) and the number of sites within the network has increased over time. Figure 2-35 compares 16 observed NH3 concentrations between 2011 and 2020. The highest observed ammonia 17 concentrations across the U.S. tend to occur in the central U.S. where values can exceed 2.4 18  $\mu$ g/m<sup>3</sup>. Consistent with expectations from the flat or slightly increasing trends in ammonia 19 emissions, we also see relatively unchanged NH<sub>3</sub> concentrations over this 10 year period, 20 although there can be some variability from site to site.



Figure 2-33. Distributions of annual mean PM<sub>2.5</sub> design values ( $\mu g/m^3$ ) at U.S. sites across the 2000-2021 period. The red line shows the number of sites included in each boxplot per year.



Distributions of the annual 98th percentile 24-hour PM2.5 design values Figure 2-34. 7  $(\mu g/m^3)$  at U.S. sites across the 2000-2021 period. The red line shows the number of sites included in each boxplot per year.

3



Figure 2-35. Annual average ammonia concentrations as measured by the Ammonia
Monitoring Network in 2010 (top) and 2020 (bottom). Data source: NADP (2012) and NADP (2021).
# 1 2.5 NITROGEN AND SULFUR DEPOSITION

2 The impacts of nitrogen and sulfur emissions on public welfare endpoints via deposition 3 are broad, complex, and variable. Contributing to the challenge of determining the impacts of 4 these pollutants are past levels of deposition of N and S, as well as other non-air related sources 5 of deposition. The focus of this review is on deposition-related impacts to ecological systems 6 from NO<sub>2</sub>, SO<sub>2</sub>, and PM. Therefore, it is important to be able to characterize deposition levels 7 across the U.S., in order to be able to understand the relationship between pollutant 8 concentrations, deposition, and subsequent adverse effects to public welfare. Assessing the 9 adequacy of any standard will require the ability to relate air quality concentrations (past and 10 present) to deposition levels (past and present). Since the previous review, the amount of N and S 11 deposition has changed, and it is important to develop the most up-to-date datasets for the 12 assessment of atmospheric deposition to capture these changes. This review assesses both 13 existing measurement data and modeling capabilities.

#### 14 2.5.1 Estimating Atmospheric Deposition

15 As introduced in Section 2.3.4, measurements of deposition are incomplete and limited. 16 While wet deposition has been routinely monitored at many locations across the U.S. for more 17 than 30 years (NADP, 2021), dry deposition is not routinely measured. As a result, most 18 deposition estimates are based on a combination of existing measurements and model 19 simulations. In 2011, the NADP established the Total Deposition (TDEP) Science Committee 20 with the goal of providing estimates of total S and N deposition across the U.S. for use in 21 estimating critical loads and other assessments. A hybrid approach has been developed to 22 estimate total deposition based on a combination of measured and modeled values, where 23 measured values are given more weight at the monitoring locations and modeled data are used to 24 fill in spatial gaps and provide information on chemical species that are not measured by routine 25 monitoring networks. One of the outputs of this effort are annual datasets of total deposition 26 estimates in the U.S. which are referred to as the TDEP datasets.

27 TDEP deposition estimates employ a combination of observations, computational 28 models, and statistical techniques (Schwede and Lear, 2014, with subsequent technical updates 29 available from NADP; ISA, Appendix 2, Section 2.6). Figure 2-36 provides a simple flowchart 30 of the process. For wet deposition, the approach is to combine the concentrations of nitrate, 31 ammonium and sulfate in precipitation as measured at NADP sites with precipitation amounts as 32 estimated in the (Parameter-elevation Relationships on Independent Slopes Model) PRISM 33 dataset. The result is a spatially complete wet deposition dataset at 4 km horizontal resolution. 34 The source of data for the dry deposition calculation is shown on the right side of Figure 2-36 35 and in more detail in Figure 2-37. Two intermediate datasets are created: an interpolated

1 measurement and a bias-corrected simulation. The interpolated measurement dataset relies on the 2 CASTNET monitoring network, which measures gas-phase SO<sub>2</sub> and nitric acid (HNO<sub>3</sub>) and particle-phase  $SO_4^{2-}$ , nitrate (NO<sub>3</sub><sup>-</sup>), and NH<sub>4</sub>. Samples are collected for one week and then 3 4 chemically analyzed. The inlet allows particles of all sizes to be collected and is designed to 5 support estimates of total oxidized nitrogen and sulfur dry deposition. Each chemical species is 6 multiplied by the effective dry deposition velocity calculated from a 12-km Community 7 Multiscale Air Quality (CMAQ) model simulation. The effective dry deposition velocity is the 8 mean dry deposition velocity over the week-long measurement. This assessment calculates the 9 effective dry deposition velocity, weighting the average by the hourly concentration, as 10 meteorological processes have an influence on both the dry deposition velocity and the 11 concentration. The result is a set of point estimates of dry deposition. These are then summed to 12 an annual total. The final step is to apply inverse distance weighted interpolation to estimate dry 13 deposition for the same 4 km horizontal resolution grid as the wet deposition dataset. 14 One shortcoming is that the measurement sites are often far apart and the TDEP 15 interpolation does not fully capture variability between the measurement locations. The TDEP 16 method calculates a bias-corrected dry deposition dataset using the results of a CMAQ 17 simulation. The bias correction is estimated by calculating the difference between the seasonal-18 average CMAQ concentrations and the CASTNET concentration measurements. The bias 19 correction at each CASTNET monitoring site is spatially interpolated to create a 4 km horizontal 20 resolution dataset. The seasonally summed CMAQ dry deposition dataset is interpolated from 21 12-km to the 4-km horizontal resolution then adjusted by the bias correction estimated from the 22 modeled and measured air concentrations. This assumes that bias in concentrations can be 23 applied to correct a bias in dry deposition, which is reasonable if the bias is due to errors in 24 emissions or chemical production but may not be true if the bias is due to inaccuracies in the dry 25 deposition rate. The four seasonally summed datasets are summed to create an annual total dry 26 deposition for each species.



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**Figure 2-36. Data sources for calculating total deposition.** Dark blue indicates observations, white boxes indicate chemical transport modeling results, and light blue boxes are the results of model-measurement fusion.



6

Figure 2-37. Data sources for estimating dry deposition. Dark blue indicates observations,
 white boxes indicate chemical transport modeling results, and light blue boxes
 are the results of model-measurement fusion. Note that the bias correction is not
 applied to ammonia, in part, because the existing method must be modified to
 account for its bidirectional flux.

#### 1 2.5.2 Uncertainty in Estimates of Atmospheric Deposition

2 Uncertainty in the resulting model-measurement fusion can be attributed to sources of 3 deposition that are not characterized by the models or measurements, uncertainties in the CMAQ 4 model results, and uncertainty in the spatially averaged deposition due to variability that is not 5 accounted for in the models. While there are multiple approaches to estimating uncertainty, this 6 review relies on what has been reported in the literature. One approach is to compare the results 7 from multiple models with similar scientific credibility. To the extent that different models 8 employ different scientific assumptions or parameterizations, this approach can give insight into 9 the scientific uncertainty. Another approach is to compare the modeling results to measurements, 10 or to withhold a subset of the data to be used as validation. This approach can provide a more 11 quantitative assessment, but it is limited by the availability of measurements. This section 12 summarizes the relevant studies that were used to provide a general assessment of uncertainty in

13 TDEP estimates of N and S deposition.

14 One source of uncertainty in the model-measurement fusion is the origin of the 15 deposition data. Some components of deposition are directly measured, some are the result of 16 combining model results and measurements, some are from modeling results only, and a small 17 fraction is not included as part of TDEP. The first step in assessing uncertainty is to assess the 18 uncertainty from each part of the TDEP calculation. Wet deposition is calculated using NADP 19 NTN nitrogen and sulfur wet deposition measurements, which are spatially interpolated and 20 combined with the PRISM estimates of precipitation. The PRISM dataset compares well with 21 NADP NTN precipitation measurements (Daly et al., 2017) and the meteorological simulations 22 from this assessment. Dry deposition relies on a combination of measurements and models and is 23 more challenging to assess. For oxidized nitrogen, air concentration of HNO<sub>3</sub> and NO<sub>3</sub><sup>-</sup> 24 particulate matter are measured at CASTNET monitoring sites. Several other compounds, such 25 as NO<sub>2</sub>, HONO, N<sub>2</sub>O<sub>5</sub>, and organic nitrogen compounds formed from photochemistry, are either 26 not routinely measured or not routinely measured in remote areas. The CMAO model estimates 27 that the deposition of the latter compounds is on average 13% of the oxidized nitrogen deposition 28 and is largest near emission sources and urban areas (Walker et al., 2019). For reduced nitrogen 29 compounds, CASTNET includes measurements of NH4<sup>+</sup> and AMoN includes measurements of 30 NH<sub>3</sub> and often these monitors are co-located. However, because of the relatively large spatial 31 variability of NH<sub>3</sub>, these ammonia measurements are not used for bias correction as part of the 32 TDEP model-measurement fusion. Dry deposition of ammonia is from the CMAQ simulation. 33 Lastly, sulfur-based compounds, SO<sub>2</sub> and particulate matter SO<sub>4</sub><sup>2-</sup> are measured at CASTNet 34 monitoring sites. Most of the largest contributors of N and S dry deposition are measured at 35 CASTNET sties which serves to constrain the modeling uncertainties. The most significant 36 exception is ammonia dry deposition, which is estimated only using CMAQ modeling results.

1 The CMAQ model is used to estimate the dry deposition velocity for chemical species 2 measured at CASTNET monitoring stations, the dry deposition in areas further from CASTNET 3 monitoring stations, and the dry deposition for species not measured by CASTNET. Like any 4 complex system, the effect of uncertainties in one model process can be reduced by 5 compensating processes. For example, consider uncertainties in the dry deposition velocity. If 6 the simulated rate of dry deposition is too high, then dry deposition would be higher in the 7 model. The enhanced dry deposition would also cause concentrations to be lower, which would 8 in turn cause wet deposition to be lower. In this case, the dry deposition would be too high, the 9 lower wet deposition would compensate for this, and the total deposition would be affected less. 10 Uncertainties that affect the rate of dry deposition relative to wet deposition will have less of an 11 effect on total deposition and can be minimized by averaging over time and space. On the other 12 hand, if the emission rates were too high, then concentrations would be higher, and both dry and 13 wet deposition would be higher. Uncertainties that affect air concentrations, such as emissions, 14 will affect both wet deposition and dry deposition, and consequently total deposition (Dennis et 15 al., 2013). Examining both air concentrations and deposition can yield insight into the nature and 16 magnitude of uncertainties in the model results.

17 Concentration measurements from CASTNET and wet deposition measurements from 18 NADP NTN are used to assess bias in the modeled deposition values. For sulfur and oxidized 19 nitrogen, the concentration and wet deposition observations are within 25% of the simulated 20 values. Because nitrate and sulfate concentrations are bias adjusted in the TDEP model-21 measurement fusion, these errors have less of an effect on the estimate of deposition in areas 22 near the measurement stations. However, in NH<sub>3</sub> concentration and NH<sub>4</sub><sup>+</sup> wet deposition bias can 23 be as high as 55%. Because the ammonia concentration and the ammonia dry deposition are not 24 constrained by measurements in the TDEP model-measurement fusion calculations, it is likely 25 that the resulting estimates for current conditions reported in this assessment overestimate 26 ammonia dry deposition due to the overestimate in ammonia concentrations. This error is most 27 pronounced in regions near large ammonia emission sources, such as confined animal feeding 28 operations (CAFOs) and fertilized crops.

29 In addition to assessing the uncertainty of the CMAQ model, it is also necessary to assess 30 the uncertainty in the NADP NTN and CASTNET measurements. The concentration and 31 deposition measurements have a specified level of precision defined in the data quality 32 objectives for each monitoring network. The NADP NTN monitors specify a less than 10% 33 uncertainty and for the CASTNET air concentration measurements the uncertainty is specified as 34 +/- 20%. This is achieved through quality assurance and data management protocols. However, 35 this may not be a complete assessment of the uncertainty. In the case of CASTNET, several 36 studies have collocated reference monitors and inter-compared the different measurement

1 techniques. Differences in sulfate tend to be small. But for nitrate and ammonium in particulate

- 2 matter, the different sampling methods can yield larger differences (ISA, Appendix 2, Section
- 3 2.4.5). The differences are thought to be increased by high humidity or influence from coastal
- 4 airmasses that affect the PM composition, and accordingly may not be relevant everywhere in
- 5 the U.S. Fully characterizing the differences that arise from different monitoring techniques is
- 6 beyond the scope of this assessment. Instead, this assessment relies on the data quality
- 7 objectives as a proxy) for uncertainty.

8 Lastly, the fusion of the model and measurements to a set spatial grid also contributes to 9 uncertainty. The grid representation of the model-measurement fusion may obscure fine 10 resolution variability leading to uncertainty in the deposition to a specific ecosystem. The dry 11 deposition velocity can differ considerably depending on the surface conditions, complex terrain, 12 elevation, and land cover. For example, the dry deposition velocity of nitric acid (HNO<sub>3</sub>) is four 13 times faster over a forest than a lake. In regions with varied terrain, this can create substantial 14 variability in the dry deposition that is not captured at the 4 km horizontal spatial scale of the 15 TDEP interpolation. This is also substantial in coastal areas or city-wildland interfaces. A study 16 by Paulot et al. (2018) estimated that grid-based results from models may underestimate 17 deposition to natural vegetation by 30%. Another issue is the spatial resolution may obscure 18 gradients in concentration. This is especially true of compounds such as NO<sub>2</sub> that have high 19 concentrations near emission sources, but degrade quickly, leading to large spatial gradients. 20 Thus, this type of uncertainty is likely less than in other, more populated areas.

21

#### 2.5.3 National Estimates of Deposition

Total sulfur and total nitrogen deposition estimates for the continental U.S. at 4-km horizontal resolution have been developed for calendar years 2000 through 2021. These data are used to quantify ecosystem effects as discussed in the later sections of this assessment. Figure 2-38 illustrates that nitrogen deposition is highest in and around large source regions. This mostly includes regions of intensive crop and animal livestock production, which are large sources of NH<sub>3</sub> emissions. The total sulfur deposition is shown in Figure 2-39. Sulfur deposition is generally higher in the eastern U.S. and near large emission sources like EGUs (section 2.2).



Figure 2-38. Three year average of the total deposition of nitrogen (kg N/ha) across the 2019-2021 period.





5 Figure 2-39. Three year average of the total deposition of sulfur (kg S/ha) across the 2019-2021 period.

#### 2.5.3.1 Contribution from NH<sub>3</sub>

Ammonia contributes to total nitrogen deposition, but it is not an oxidized form of nitrogen, so it is not part of the definition of "oxides of nitrogen". In addition, although ammonia is a precursor to PM formation, ammonia is a gas and not a component of particulate matter. Accordingly, ammonia is not specifically within in the scope of the criteria pollutants that are part of this review, and therefore it is necessary to quantify the contribution of ammonia to nitrogen deposition separately from the other components of nitrogen deposition.

8 This review applies the CMAQ model with additional enhancements to track the 9 contribution of ammonia to both dry and wet deposition. First, for dry deposition, the CMAQ 10 model separately tracks the each of the main chemical species that include nitrogen, including 11 ammonia. This is important, because each of the chemical species has a different dry deposition 12 velocity, depending on that compound's physical properties. For wet deposition, CMAQ uses an 13 equilibrium approach. Based on the temperature, relative humidity, and relative concentration of 14 particle and gas-phase concentrations, CMAQ calculates the pH of the cloud droplets as well as 15 the equilibrium concentration of each species in the cloud water, in particle form, and in the gas 16 phase. The most thermodynamically favorable state is for nearly all the ammonia in the cloud 17 droplet to be in the form of ammonium ion (NH<sub>4</sub><sup>+</sup>). From the model results alone, we would 18 attribute nearly all the wet deposition to be in the form of ammonium, rather than ammonia. 19 However, much of the nitrogen that enters the cloud droplet is in the gas-phase as ammonia. In 20 CMAQ, the contribution of ammonia to the cloud droplet ammonium is accounted for by taking 21 the difference between the gas-phase concentration of ammonia before the cloud and after the 22 cloud equilibrium calculation. This portion from ammonia is tracked in a separate variable. It 23 does not change the model calculations in any way; it is just used to account for the contribution 24 of ammonia to wet deposition of N. 25 The contribution of ammonia to total nitrogen deposition, as averaged over 2019 - 2021, 26 is shown in Figure 2-40. Deposition of ammonia is calculated as the sum of dry deposition of 27 ammonia and wet deposition of ammonia as described above. Total nitrogen deposition is the 28 sum of ammonia, ammonium, and oxidized nitrogen compounds. The contribution of ammonia 29 exceeds 70% in areas with large ammonia emissions, including areas of intensive livestock and

30 crops production in eastern North Carolina, parts of Iowa, Minnesota, Texas, and the Central and

- 31 Imperial valleys in California.
- 32



Figure 2-40. Average percent of total N deposition in 2019-2021 as reduced N (gas phase NH<sub>3</sub> and particle phase NH<sub>4</sub><sup>+</sup>)

#### 2.5.3.2 Contribution from International Transport

5 On a national average scale, only a small fraction of sulfur and nitrogen deposition can be 6 attributed to natural emissions or international transport (ISA, Appendix 2, Section 2.6.8). 7 Chemical transport models have been used to quantify these contributions (Horowitz et al., 2003; 8 Zhang et al., 2012; Lee et al., 2016). The natural sources of oxidized nitrogen include 9 unfertilized soils and lightning. Ammonia is emitted from unfertilized soils and from wild 10 animals. Chemical transport model simulations have been used to estimate that natural emission 11 sources contribute 16% of the total N deposition in the U.S. Because ammonia and most forms of 12 oxidized N have relatively short atmospheric lifetimes, international transport contributes just 13 6% of the N deposition, except within 100 km of the U.S.-Canada or U.S.-Mexico borders, 14 where the contribution is estimated to be at most 20%. U.S. anthropogenic emissions account for 15 78% of Nr deposition over the contiguous United States (CONUS) (ISA, Appendix 2, Section 16 2.6.8). Sulfur is naturally emitted from plankton in the ocean and from geologic activity –

17 volcanoes, fumaroles, etc. Like N, relatively little sulfur deposition can be attributed to

1 2

3

1 international transport. Chemical transport model simulations have been used to estimate that

2 approximately 10% of S in the eastern U.S. can be attributed to natural and international sources.

3 In the western U.S., this increases to 20%, since there is lower S deposition from anthropogenic

4 sources, more geologic emission sources, and closer proximity to long range transport from

5 international sources. In areas with high S deposition, less than 1% can be attributed to natural

6 and international sources (ISA, Appendix 2, Section 2.6.8).

#### 7 2.5.4 Trends in Deposition

8 With the changes in emissions and air concentrations described above, total deposition of 9 oxidized nitrogen and sulfur have also decreased significantly since 2000 (Feng et al., 2020; 10 McHale et al., 2021). Between the three-year period 2000-2002 and 2018-2020, national average 11 for CONUS S deposition has declined by 68% and total N deposition has declined by 15% (U.S. 12 EPA, 2022b). See Tables 2-2a and 2-2b for a regional breakout of trends in total S, total N, 13 oxidized N, and reduced N deposition trends. The change in total N deposition is a combination 14 of declining oxidized N and increasing reduced N, which is similar to the trend in emissions and 15 air concentrations described above. Emissions of NO<sub>X</sub> and wet deposition of nitrate have a 16 positive correlation, but because the formation of ammonium is related to the availability of 17 nitrate and sulfate, the correlation between  $NH_3$  emissions and  $NH_4^+$  wet deposition is weaker 18 and negative (Tan et al., 2020). While dry deposition is more uncertain in magnitude, both 19 surface-based and remote-sensing measurements indicate increasing ammonia concentrations, 20 which points to an increasing trend for ammonia dry deposition, especially in areas with 21 significant agricultural emissions in the Midwest and Central Valley of California where 22 ammonia dry deposition has become the largest contributor to inorganic N deposition (Li et al., 23 2016). Figure 2-41 shows HNO<sub>3</sub> ambient concentration data for a past and recent year (1996 and 24 2019) and then Figure 2-42 displays how those changes in concentrations have translated to 25 changes in model-estimated HNO<sub>3</sub> dry deposition over similar time periods. As expected, the 26 data suggest that dry deposition of nitric acid has decreased significantly over the past two 27 decades and is likely a key contributor to the decrease in total nitrate deposition and decreasing 28 trends in oxidized nitrogen deposition (ISA, Appendix 2, Section 2.7). 29

# Table 2-2.Change in total deposition by region between the 2000-2002 and 2019-2021<br/>periods (U.S. EPA, 2022b): (a) total S deposition; (b) total, oxidized and<br/>reduced N deposition.

Form of S Deposition	Region	2000-2002	2019-2021	% change
	Mid-Atlantic	15.9	2.1	-87
	Midwest	11.2	2.2	-80
	North Central	3.5	1.5	-56
Total Deposition of Sulfur (kg S ha <sup>_1</sup> )	Northeast	8.7	1.5	-83
	Pacific	1.0	0.6	-38
	Rocky Mountain	1.0	0.6	-46
	South Central	5.4	2.8	-49
	Southeast	10.3	2.6	-74

#### (a) Change in total S deposition

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2 3

#### (b) Change in total, oxidized and reduced N deposition

Form of N Deposition	Region	2000-2002	2019-2021	% change
Total Deposition of Nitrogen (kg N ha <sup>-1</sup> )	Mid-Atlantic	13.4	8.5	-36
	Midwest	12.2	9.8	-20
	North Central	8.5	9.5	+11
	Northeast	10.4	6.2	-40
	Pacific	3.8	3.1	-18
	Rocky Mountain	3.0	3.1	+3
	South Central	7.8	9.0	+16
	Southeast	10.8	8.4	-23
Total Deposition of Oxidized Nitrogen (kg N ha <sup>-1</sup> )	Mid-Atlantic	10.3	4.0	-62
	Midwest	8.0	3.6	-54
	North Central	4.1	2.6	-37
	Northeast	7.7	2.9	-62
	Pacific	2.4	1.4	-42
	Rocky Mountain	1.9	1.3	-35
	South Central	5.0	3.1	-39
	Southeast	7.7	3.4	-56
Total Deposition of Reduced Nitrogen (kg N ha <sup>-1</sup> )	Mid-Atlantic	3.0	4.6	+51
	Midwest	4.3	6.2	+45
	North Central	4.4	6.9	+56
	Northeast	2.7	3.3	+22
	Pacific	1.4	1.7	+22
	Rocky Mountain	1.1	1.8	+72
	South Central	2.8	6.0	+111
	Southeast	3.1	5.0	+63









1 The trends in deposition of reduced nitrogen should be viewed with some caution, in part 2 because before 2011, ambient air NH<sub>3</sub> monitoring was rare. For particulate matter, the trend in 3 ammonium (NH<sub>4</sub><sup>+</sup>) has followed the downward trends in sulfate and nitrate, because in order for 4 NH<sub>3</sub> to partition into the particle phase, an anion, such as sulfate or nitrate, is needed to 5 neutralize it. Satellite-based measurements and chemical transport models have been used to 6 augment the surface-based measurements of ammonia and ammonium to better understand 7 trends. These studies also show increasing ammonia concentrations, especially in parts of the 8 Midwest, South-east, and West near agricultural sources (Warner et al., 2016; Warner et al., 9 2017; Yu et al., 2018; Nair et al., 2019; He et al., 2021). These trends are attributed to a 10 combination of warmer temperatures causing greater emissions, increasing agricultural activity, 11 and less available sulfate and nitrate, shifting particle ammonium to gas-phase ammonia. 12 While there is always uncertainty in projecting future trends, the EPA generally expects 13 reductions in total national N and S deposition over the next decade, although trends in reduced 14 N deposition will remain a concern. In a recent regulatory impact assessment for the proposed 15 revisions to the PM NAAQS, the EPA used the CMAQ model to simulate an illustrative 16 implementation scenario that included additional emissions reductions of  $NO_X$  and  $SO_2$  (U.S. 17 EPA, 2022a) The percent change in total N and total S deposition projected to occur by the model in 2032 (from a baseline 2016 scenario) within Class 1 areas is shown in Figure 2-43 and 18 19 Figure 2-44, respectively. In this scenario, deposition in Class I Areas is expected to continue to 20 decline as existing regulations are implemented, due to reductions in NO<sub>X</sub> and SO<sub>2</sub> emissions. 21 The projected average deposition reduction for N and S is about 10%, with largest reductions 22 occurring in the East. The projected reduction in sulfur in the Pacific Coast states is relatively 23 minor, but there is already very little sulfur deposition and very few  $SO_2$  emission sources in this 24 region. Areas with relatively high levels of deposition in 2016 have the largest projected 25 reduction in deposition, but reductions in deposition are not limited to just these high deposition 26 areas, with deposition at nearly all Class I Areas expected to decline further. It should be noted 27 that there is considerable uncertainty in the change in future deposition due to the any revision to 28 the annual average PM<sub>2.5</sub> primary standard. The emission sources that typically contribute most 29 to high PM<sub>2.5</sub> concentrations can be located relatively far from more remote Class I Areas and 30 can have a highly variable effect on deposition in those areas. Second, as part of implementation, 31 States could elect to reduce emission sources that contribute to organic carbon  $PM_{2.5}$  which have 32 little impact on deposition.

N Change in Deposition scenario minus base case



Figure 2-43. Projected percent change in total N deposition in Class 1 areas from 2016, based on a scenario for 2032 that includes implementation of existing national rules on mobile and stationary sources (U.S. EPA, 2022a).



Figure 2-44. Projected percent change in total S deposition in Class 1 areas from 2016, based on a scenario for 2032 that includes implementation of existing national rules on mobile and stationary sources (U.S. EPA, 2022a).

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# **3** THE CURRENT STANDARDS AND GENERAL APPROACH FOR THIS REVIEW

3 This review focuses on evaluation of the currently available evidence and quantitative 4 analyses related to the welfare effects of oxides of S and N and the ecological effects of PM in 5 consideration of several overarching policy-relevant questions. The first such question considers whether the currently available scientific evidence and quantitative information support or call 6 7 into question the adequacy of the public welfare protection for these effects afforded by the 8 current secondary standards for these pollutants. In this context we consider two categories of 9 effects: (1) effects associated with the airborne pollutants (sometimes referred to as "direct 10 effects"), and (2) effects associated with deposition of the pollutants or their transformation products into aquatic and terrestrial ecosystems. 11 12 This chapter describes the basis for the existing secondary standards (section 3.1) and the

approach taken in the 2012 review of deposition-related effects (section 3.2) and outlines the
approach being taken in this review of the current NO<sub>2</sub>, SO<sub>2</sub> and PM secondary standards
(section 3.3).

# 16 3.1 BASIS FOR THE EXISTING SECONDARY STANDARDS

17 The existing secondary standards for oxides of S and N were established in 1971 (36 FR 18 8186, April 30, 1971). The secondary standard for SO<sub>2</sub> is 0.5 ppm, as a 3-hour average, not to be 19 exceeded more than once per year (40 CFR §50.5). The secondary standard for N oxides is 0.053 20 ppm NO<sub>2</sub> (100 micrograms per cubic meter [ $\mu$ g/m<sup>3</sup>] of air), as the arithmetic mean of the 1-hour 21 NO<sub>2</sub> concentrations over the course of a year (40 CFR §50.11). Both standards were selected to 22 provide protection to the public welfare related to effects on vegetation.

23 The welfare effects evidence for  $SO_X$  in previous reviews indicates a relationship 24 between short- and long-term SO<sub>2</sub> exposures and foliar damage to cultivated plants, reductions in 25 productivity, species richness, and diversity (U.S. EPA, 1969; U.S. EPA, 1982; U.S. EPA, 2008). 26 At the time the standard was set, concentrations of SO<sub>2</sub> in the ambient air were also associated 27 with other welfare effects, including effects on materials, visibility, soils, and water. However, 28 the available data were not sufficient to establish a quantitative relationship between specific  $SO_2$ 29 concentrations and such effects (38 FR 25679, September 14, 1973). Accordingly, the existing 30 secondary standard for SO<sub>X</sub> was established with a focus on providing public welfare protection 31 related to the direct effects on vegetation of SO<sub>X</sub> in ambient air. 32 The welfare effects evidence for N oxides in previous reviews includes foliar injury, leaf 33 drop and reduced yield of some crops (U.S. EPA, 1971; U.S. EPA, 1982; U.S. EPA, 1993; U.S.

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1 EPA, 2008). Since it was established in 1971, the secondary standard for N oxides has been

2 reviewed three times, in 1985, 1996, and 2012. Although those reviews identified additional

3 effects related to N deposition, they concluded that the existing standard provided adequate

4 protection related to the vegetation effects of airborne N oxides (i.e., the "direct" effects of N

5 oxides in ambient air).

6 The existing secondary standards for PM, include two PM<sub>2.5</sub> standards and one PM<sub>10</sub> 7 standard. The PM<sub>2.5</sub> standards are 35  $ug/m^3$  as the average of three consecutive annual 98<sup>th</sup> percentile 24-hour averages and 15.0 ug/m<sup>3</sup>, as an annual mean concentration, averaged over 8 9 three years (40 CFR §50.13). The PM<sub>10</sub> standard is 150 ug/m<sup>3</sup> as a 24-hour average, not to be 10 exceeded more than once per year on average over three years (40 CFR §50.6). These standards 11 address an array of effects that include effects on visibility, materials damage, and climate 12 effects, as well as ecological effects, including those related to deposition. It is only the latter – 13 ecological effects, including those related to deposition - that fall into this review. The existing 14 PM secondary standards have not generally been established with ecological effects as their 15 focus, although prior reviews have generally concluded them to provide protection for such

16 effects (e.g., 78 FR 3086, January 15, 2013).

#### **3.2 PRIOR REVIEW OF DEPOSITION-RELATED EFFECTS**

18 The most recent review of the NO<sub>2</sub> and SO<sub>2</sub> secondary standards was completed in 2012. 19 In that review, the EPA recognized that a significant increase in understanding of the effects of 20 oxides of nitrogen and sulfur had occurred since the prior secondary standards reviews for those 21 pollutants, reflecting the large amount of research that had been conducted on the effects of 22 deposition of nitrogen and sulfur to ecosystems (77 FR 20236, April 3, 2012). Considering the 23 extensive evidence available at that time, the Agency concluded that the most significant current 24 risks of adverse effects to public welfare associated with those pollutants are those related to 25 deposition of oxides of nitrogen and sulfur to both terrestrial and aquatic ecosystems (77 FR 26 20236, April 3, 2012). Accordingly, in addition to evaluating the protection provided by the 27 secondary standards for oxides of nitrogen and oxides of sulfur from effects associated with the 28 airborne pollutants, the 2012 review also included extensive analyses of the welfare effects 29 associated with nitrogen and sulfur deposition to sensitive aquatic and terrestrial ecosystems (77 30 FR 20218, April 3, 2012).

Based on the available evidence, the risks of atmospheric deposition analyzed in the 2009 REA related to two categories of ecosystem effects, acidification and nutrient enrichment. The analyses included assessment of risks of both types of effects in both terrestrial and aquatic ecosystems. While the available evidence supported conclusions regarding the role of atmospheric deposition of oxides of N and S in acidification and nutrient enrichment of aquatic 1 and terrestrial ecosystems, there was variation in the strength of the evidence and of the

- 2 information supporting the multiple quantitative linkages between the pollutants in ambient air
- 3 and responses of terrestrial and aquatic ecosystems, their associated biota, and potential public
- 4 welfare implications. As a result, the focus in the 2012 review with regard to consideration of a
- 5 secondary standard to provide protection from deposition-related effects of oxides of N and S
- 6 was on the information related to aquatic acidification (U.S. EPA, 2011, chapter 7).

7 With regard to acidification-related effects in terrestrial ecosystems, the 2009 REA had 8 analyzed risks to sensitive tree species in the northeastern U.S. using the ecological indicator, 9 soil BC:Al (base cations to aluminum) ratio, which has links to tree health and growth. While the 10 analyses indicated results of potential concern with regard to 2002 levels of acid deposition, 11 several uncertainties affected the strength of associated conclusions. As noted in the 2012 12 decision, an important drawback in understanding terrestrial acidification is related to the 13 sparseness of available data for identifying appropriate BC:Al ratio target levels, and that the 14 then-available data were based on laboratory responses rather than on field measurements (77 FR 15 20229, April 3, 2012). The 2012 decision also recognized uncertainties with regard to empirical 16 case studies in the ISA noting that other stressors present in the field that are not present in the 17 laboratory may confound the relationship between N oxides and SO<sub>X</sub> deposition and terrestrial 18 acidification effects (U.S. EPA, 2008, section 3.2.2.1 and 77 FR 20229, April 3, 2012). The REA 19 analyses of aquatic acidification (which involved water quality modeling of acid deposition in 20 case study watersheds and prediction of waterbody acid neutralizing capacity (ANC) response), 21 however, provided strong support to the evidence for a relationship between atmospheric 22 deposition of oxides of nitrogen and sulfur and loss of acid neutralizing capacity in sensitive 23 ecosystems, with associated aquatic acidification effects.

24 Consideration of the nutrient enrichment-related effects of atmospheric N and S 25 deposition with regard to identification of options to provide protection for deposition-related 26 effects was limited by several factors. For example, while there is extensive evidence of 27 deleterious effects of excessive nitrogen loadings to terrestrial ecosystems, the co-stressors 28 affecting forests, including other air pollutants such as ozone, and limiting factors such as 29 moisture and other nutrients, confound the assessment of marginal changes in any one stressor or 30 nutrient in a forest ecosystem, leaving the information on the effects of changes in N deposition 31 on forestlands and other terrestrial ecosystems limited (U.S. EPA, 2011, section 6.3.2). Further, 32 the 2008 ISA noted that only a fraction of the deposited nitrogen is taken up by the forests, with 33 most of the nitrogen retained in the soils (U.S. EPA, 2008, section 3.3.2.1), and that forest 34 management practices can significantly affect the nitrogen cycling within a forest ecosystem. 35 Accordingly, the response of managed forests to N oxides deposition will be variable depending 36 on the forest management practices employed in a given forest ecosystem (U.S. EPA, 2008,

1 Annex C, section C.6.3). Factors affecting consideration of aquatic eutrophication effects

- 2 included the appreciable contributions of non-atmospheric sources to waterbody nutrient loading
- 3 which affected our attribution of specific effects to atmospheric sources of N, and limitations in
- 4 the ability of the available data and models to characterize incremental adverse impacts of N
- 5 deposition (U.S. EPA, 2011, section 6.3.2).

6 Thus, in light of the evidence and findings of these analyses, and advice from the 7 CASAC, the EPA concluded it had the greatest confidence in findings related to the aquatic 8 acidification-related effects of oxides of nitrogen and sulfur relative to other deposition-related 9 effects. Therefore, the PA focused on aquatic acidification effects from deposition of nitrogen 10 and sulfur in identifying policy options for providing public welfare protection from deposition-11 related effects of oxides of N and S, concluding that the available information and assessments 12 were only sufficient at that time to support development of a standard to address aquatic 13 acidification. Consistent with this, the PA concluded it was appropriate to consider a secondary 14 standard in the form of an aquatic acidification index (AAI) and identified a range of AAI values 15 (which correspond to minimum ANC levels) for consideration (U.S. EPA, 2011, section 7.6.2).

16 Conceptually, the AAI is an index that utilizes the results of ecosystem and air quality 17 modeling to estimate waterbody ANC. Thus, the standard level for an AAI-based standard is a national minimum target ANC for waterbodies in the ecoregions of the U.S. for which the data 18 19 are considered adequate for these purposes. While the NAAQS have historically been set in 20 terms of an ambient atmospheric concentration or mixing ration, an AAI-based standard was 21 envisioned to have a single value established for the AAI, but the concentrations of SO<sub>X</sub> and N 22 oxides would be specific to each ecoregion, taking into account variation in several factors that 23 influence waterbody ANC, and consequently could vary across the U.S. The factors, specific to 24 each ecoregion, which it was envisioned would be established as part of the standard, include: 25 surface water runoff rates and so-called "transference ratios," which are factors applied to back-26 calculate or estimate the concentrations of SO<sub>X</sub> and N oxides corresponding to target deposition 27 values that would meet the AAI-based standard level, which is also the target minimum ANC 28 (U.S. EPA, 2011, chapter 7).<sup>1</sup> The ecoregion-specific values for these factors would be specified 29 based on then available data and simulations of the CMAQ model, and codified as part of such a 30 standard. As part of the standard, these factors would be reviewed in the context of each periodic 31 review of the NAAOS.

32 After consideration of the PA conclusions, the Administrator concluded that while the 33 conceptual basis for the AAI was supported by the available scientific information, there were

<sup>&</sup>lt;sup>1</sup> These were among the ecoregion-specific factors that comprised the parameters, F1 through F4 in the AAI equation (2011 PA, p. 7-37).

1 limitations in the available relevant data, and uncertainties associated with specifying the

- 2 elements of the AAI, specifically those based on modeled factors, that posed obstacles to
- 3 establishing such a standard under the Clean Air Act. In so doing, it was recognized that the
- 4 general structure of an AAI-based standard addressed the potential for contributions to acid
- 5 deposition from both oxides of nitrogen and of sulfur, and quantitatively described linkages
- 6 between ambient concentrations, deposition, and aquatic acidification, considering variations in
- 7 factors affecting in these linkages across the country. However, the limitations and uncertainties
- 8 in the available information were judged to be too great to support establishment of a new
- 9 standard that could be concluded to provide the requisite protection for such effects under the
- 10 Act (77 FR 20218, April 3, 2012). The Administrator concluded that while the current
- 11 secondary standards were not adequate to provide protection against potentially adverse
- 12 deposition-related effects associated with oxides of nitrogen and sulfur, it was not appropriate
- 13 under Section 109 to set any new or additional standards at that time to address effects associated
- 14 with deposition of oxides of nitrogen and sulfur on sensitive aquatic and terrestrial ecosystems
- 15 (77 FR 20218, April 3, 2012).

# 16 **3.3 GENERAL APPROACH FOR THIS REVIEW**

17 As is the case for all NAAQS reviews, this secondary standards review is fundamentally 18 based on using the Agency's assessment of the current scientific evidence and associated 19 quantitative analyses to inform the Administrator's judgments regarding secondary standards that 20 are requisite to protect the public welfare from known or anticipated adverse effects. The 21 approach planned for this review of the secondary N oxides, SO<sub>X</sub>, and PM standards will build 22 on the last reviews, including the substantial assessments and evaluations performed over the 23 course of those reviews, and considering the more recent scientific information and air quality 24 data now available to inform understanding of the key policy-relevant issues in the current 25 review.

26 The evaluations in the PA, including the scientific assessments in the ISA (building on 27 prior such assessments) augmented by quantitative air quality and exposure analyses, are 28 intended to inform the Administrator's public welfare policy judgments and conclusions, 29 including his decisions as to whether to retain or revise the standards. The PA considers the 30 potential implications of various aspects of the scientific evidence, the air quality, exposure or 31 risk-based information, and the associated uncertainties and limitations. In so doing, the 32 approach for this PA involves evaluating the available scientific and technical information to 33 address a series of key policy-relevant questions using both evidence- and exposure/risk-based 34 considerations. Together, consideration of the full set of evidence and information available in 35 this review will inform the answer to the following initial overarching question for the review:

Do the currently available scientific evidence and exposure-/risk-based information
 support or call into question the adequacy of the public welfare protection afforded by
 the current secondary standards?

4 In reflecting on this question in Chapter 7 of this PA, we consider the available body of 5 scientific evidence, assessed in the ISA (summarized in Chapters 4 and 5), and considered as a 6 basis for developing or interpreting the quantitative information, including air quality and 7 exposure analyses (summarized in Chapters 5 and 6), including whether it supports or calls into 8 question the scientific conclusions reached in the last review regarding welfare effects related to 9 SO<sub>X</sub>, N oxides and PM in ambient air. Information available in this review that may be 10 informative to public policy judgments on the significance or adversity of key effects on the 11 public welfare is also considered. Additionally, the currently available exposure and risk 12 information, whether newly developed in this review or predominantly developed in the past and 13 interpreted in light of current information, is considered. Further, in considering this question 14 with regard to these secondary standards, we give particular attention to exposures and risks for 15 effects with the greatest potential for public welfare significance. 16 The approach to reaching conclusions on the current secondary standards and, as 17 appropriate, on potential alternative standards, including consideration of policy-relevant

18 questions that frame the current review, is illustrated in Figure 3-1.





Figure 3-1. Overview of general approach for review of the secondary N oxides, SOx, and
 PM standards.

1 The Agency's approach in its review of secondary standards is consistent with the 2 requirements of the provisions of the CAA related to the review of NAAQS and with how the 3 EPA and the courts have historically interpreted the CAA. As discussed in section 1.2 above, 4 these provisions require the Administrator to establish secondary standards that, in the 5 Administrator's judgment, are requisite (i.e., neither more nor less stringent than necessary) to 6 protect the public welfare from known or anticipated adverse effects associated with the presence 7 of the pollutant in the ambient air. In so doing, the Administrator considers advice from the 8 CASAC and public comment.

9 Consistent with the Agency's approach across all NAAQS reviews, the approach of this 10 PA informs the Administrator's judgments based on a recognition that the available welfare 11 effects evidence generally reflects a range of effects that include ambient air exposure 12 circumstances for which scientists generally agree that effects are likely to occur as well as lower 13 levels at which the likelihood and magnitude of response become increasingly uncertain. The 14 CAA does not require that standards be set at a zero-risk level, but rather at a level that reduces 15 risk sufficiently so as to protect the public welfare from known or anticipated adverse effects.

16 The Agency's decisions on the adequacy of the current secondary standards and, as 17 appropriate, on any potential alternative standards considered in a review, are largely public 18 welfare policy judgments made by the Administrator. The four basic elements of the NAAQS 19 (i.e., indicator, averaging time, form, and level) are considered collectively in evaluating the 20 protection afforded by the current standard, or any alternative standards considered. Thus, the 21 Administrator's final decisions in such reviews draw upon the scientific information and 22 analyses about welfare effects, environmental exposures and risks, and associated public welfare 23 significance, as well as judgments about how to consider the range and magnitude of 24 uncertainties that are inherent in the scientific evidence and quantitative analyses.

#### 25

### 3.3.1 Approach for Direct Effects of the Pollutants in Ambient Air

26 As in past reviews of secondary standards for SO<sub>X</sub>, N oxides and PM, this review will 27 continue to assess the protection provided by the standards from effects of the airborne 28 pollutants. Accordingly, this PA draws on the currently available evidence as assessed in the 29 ISA, including the determinations regarding the causal nature of relationships between the 30 airborne pollutants and ecological effects, which focus most prominently on vegetation, and 31 quantitative exposure and air quality information (summarized in Chapters 4 and 5). Based on 32 this information, we will consider the policy implications, most specifically in addressing the 33 overarching questions articulated in section 3.3 above. Building from these considerations, the 34 PA will preliminarily conclude whether the evidence supports the retention or revision of the 35 current NO<sub>2</sub> and SO<sub>2</sub> secondary standards. With regard to the effects of PM, we will take a

- 1 similar approach, based on the evidence presented in the current ISA and conclusions from the
- 2 2012 review of the PM NAAQS (in which ecological effects were last considered) to assess the
- 3 effectiveness of the current PM standard to protect against these types of impacts.

#### 4 3.3.2 Approach for Deposition-Related Ecological Effects

5 In addition to evaluating the standards as to protection for effects of the airborne 6 pollutants, we are also evaluating the standards as to protection from deposition-related effects. 7 In so doing, we have considered the quantitative analyses conducted in the last review of the 8 relationships between oxides of nitrogen and oxides of sulfur and deposition related effects and 9 considerations for secondary standards. The overall approach we are employing takes into 10 account the nature of the welfare effects and the exposure conditions associated with effects in 11 order to identify deposition-level benchmarks appropriate to consider in the context of public 12 welfare protection. To identify metrics relevant to air quality standards (and their elements), we 13 apply relationships developed from air quality measurements near pollutant sources and 14 deposition estimates in sensitive ecoregions. From these, we identify an array of policy options 15 that might be expected to provide protection from adverse effects to the public welfare. This 16 approach is illustrated in Figure 3-2 below.



- Figure 3-2. General approach for assessing the currently available information with
   regard to consideration of protection provided for deposition-related
   ecological effects on the public welfare.
- 21 Our consideration of the nature of the welfare effects draws on the overview provided in
- 22 Chapter 4, based on the evidence presented in the ISA, key limitations in this evidence, and the
- 23 associated uncertainties. These effects encompass both effects of airborne N oxides and SO<sub>X</sub>, as

1 well as deposition-related effects, including terrestrial and aquatic acidification effects, as well as

2 effects from N enrichment. In so doing, we take note of the public welfare implications

3 associated with such effects (as summarized in section 4.3).

4 Next, we consider the current information on exposure conditions associated with effects 5 (Chapter 5) in order to identify deposition levels appropriate to consider in the context of public 6 welfare protection. We investigate the extent to which the available evidence provides 7 quantitative information linking N oxides, SO<sub>X</sub>, and PM to deposition-related effects that can 8 inform judgements on the likelihood of occurrence of such effects in air quality that meets the 9 current standard. In critically assessing the available quantitative information, we recognize that 10 the impacts of N and S deposition, which include ecosystem acidification and nutrient enrichment, are influenced by past deposition. The historical deposition associated with oxides of 11 12 S and N and PM in ambient air has modified soil and waterbody chemistry with associated 13 impacts on terrestrial and aquatic ecosystems and organisms (U.S. EPA, 2020; U.S. EPA, 2008; U.S. EPA, 1982)<sup>2</sup>. 14 15 These impacts from the dramatically higher deposition of the past century can affect how 16 ecosystems and biota respond to more recent lower deposition rates, complicating interpretation 17 of impacts related to more recent, lower deposition levels. This complexity is illustrated by findings of some studies that compared soil chemistry across 15-30-year intervals (1984-2001 18 19 and 1967-1997) and reported that although atmospheric deposition in the Northeast declined 20 across those intervals, soil acidity increased (ISA, section 4.6.1). As noted in the ISA, "[i]n areas 21 where N and S deposition has decreased, chemical recovery must first create physical and 22 chemical conditions favorable for growth, survival, and reproduction" (ISA, section 4.6.1). Thus, 23 the extent to which S and N compounds are retained in soil matrices, once deposited, with 24 potential effects on soil chemistry, as well as ambient air concentrations and associated

25 deposition, influence the dynamics of the response of the various environmental pathway

deposition, influence the dynamics of the response of the various environmental pathways tochanges in air quality.

Based on the information summarized in Chapter 5 for aquatic and terrestrial systems, we
seek to identify deposition levels associated with welfare effects of potential concern for
consideration with regard to secondary standard protection. In so doing, one objective is to
discern for what effects the evidence is most robust with regard to established quantitative

<sup>&</sup>lt;sup>2</sup> The role of historical deposition in current ecosystem circumstances (e.g., waterbody acidification and loss of aquatic species, terrestrial acidification, and aquatic eutrophication) and the complications affecting recovery have been noted in scientific assessments for NAAQS reviews ranging from the 1982 AQCD for PM and SO<sub>X</sub> to the current ISA (ISA, sections IS.2.3, IS.5.1.2, IS.6.1.1.1, and IS.11, Appendix 4, section 4.8.5, Appendix 6, section 6.6.3, Appendix 7, sections 7.1.5, 7.1.7, and 7.2.7, Appendix 8, sections 8.3.1.1, 8.4.1, 8.4.4, 8.4.5 8.6.6, and 8.6.8, Appendix 9, 9.3.2.1, Appendix 10, section 10.2.5, Appendix 12, section 12.3.3.4; 2008 ISA, sections 3.2.1.2, 3.2.3, 3.2.4.3 and 3.2.4.4; 1982 AQCD, section 1.7 and Chapter 7).

1 relationships between deposition and ecosystem effects. In this context, we present an analysis of

- 2 the findings in the currently available evidence, as well as additional quantitative analyses as
- 3 they relate to effects of airborne N oxides, SO<sub>X</sub>, and PM and deposition-related effects. The
- 4 information for terrestrial ecosystems is derived primarily from analysis of the evidence
- 5 presented in the ISA. For aquatic ecosystems, we give primary focus to aquatic acidification, for
- 6 which we have conducted quantitative analyses (based on steady-state water quality modeling) to
- 7 describe the relationships between acid deposition and acid neutralizing capacity in U.S.
- 8 ecoregions.

9 In a parallel track, we have utilized air quality modeling to characterize atmospheric 10 transport of the pollutants from their occurrence at monitors near their point of release to distant 11 ecoregions where they might be expected to deposit (Chapter 6). Based on these tracks which 12 inform an understanding of the relative contributions of source locations to individual ecoregions 13 in the U.S., we develop quantitative relationships of air pollutant concentrations to atmospheric 14 deposition rates. To identify metrics relevant to air quality standards (and their elements), we 15 apply relationships developed from air quality measurements near pollutant sources and 16 deposition estimates in sensitive ecoregions. This will consider existing standard metrics, as well 17 as other potential metrics for effective deposition-related standards. In so doing, we also 18 recognize key uncertainties and limitations in relating deposition to measurements of air quality, 19 as well as uncertainties and limitations associated with various exposure metrics. Thus, in 20 combination with the identified deposition levels of interest, we consider the extent to which 21 existing standards provide protection from these levels and seek to identify potential alternative 22 standards that might afford such protection (Chapter 7). Based on these considerations we 23 identify an array of policy options for consideration in this review

24 **3.3.3** Identification of Policy Options

25 When final, this PA is intended to provide a range of potential policy options, supported 26 by the science, to inform the Administrator's decisions regarding secondary standards that 27 provide the "requisite" public welfare protection from these pollutants in ambient air. In so 28 doing, this PA considers the evidence and quantitative analyses for direct effects of the pollutants 29 in ambient air as well as the effects of the pollutants deposited into aquatic and terrestrial 30 ecosystems, as described in sections 3.3.1 and 3.3.2 above, with regard to the policy-relevant 31 questions identified for the review. Based on those considerations (discussed in Chapter 7), we 32 consider the overarching questions for the review with regard to the extent to which the current 33 information calls into question any of the existing standards, and the extent to which new or 34 revised standards are appropriate to consider. Considerations are discussed and conclusions

reached with regard to protection from effects of the airborne pollutants and deposition-related
 effects.

3 In considering potential alternative standards, as appropriate, we evaluate what the 4 current information, including emissions and air quality analyses available in Chapter 2, may 5 indicate regarding the relationships between N oxides, SO<sub>X</sub>, and PM and N/S deposition, the 6 influence of different averaging times on N/S deposition, and what the quantitative analyses 7 indicate regarding the extent to which one or more standards may have the potential for 8 controlling deposition-related and other effects of concern (Chapter 7). In so doing, we consider 9 potential alternative standards of the same indicator and averaging time as existing standards, as 10 well as options involving different averaging times and/or indicators, in order to inform the 11 Administrator's judgements on the currently available information and what the available 12 information indicates regarding what control of air quality (and as appropriate, associated 13 deposition) may be exerted by alternative standards. Finally, the PA will present the staff 14 preliminary conclusions on whether the current evidence and quantitative analyses call into 15 question the adequacy of protection from ecological effects afforded by the SO<sub>2</sub>, NO<sub>2</sub>, and PM 16 secondary standards, and what alternative standards may be appropriate for the Administrator to 17 consider. 18 In identifying policy options appropriate to consider for providing protection from 19 deposition-related effects, we are mindful of the long history of greater and more widespread

20 atmospheric emissions that occurred in previous years (both before and after establishment of the

21 existing NAAQS) and that has contributed to acidification and/or nutrient enrichment of aquatic

and terrestrial ecosystems, the impacts of which exist to some extent in some ecosystems today.

23 This historical backdrop additionally complicates policy considerations related to deposition-

24 related effects and the identification of appropriate targets for protection in ecosystems today that

25 might be expected to protect key ecosystem functions in the context of changing conditions over

time.

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# **4 NATURE OF WELFARE EFFECTS**

2 In this chapter we summarize the current evidence on the ecosystem effects of oxides of 3 nitrogen, oxides of sulfur and particulate matter in ambient air. We consider both the evidence 4 for direct effects of the pollutants in ambient air and for the effects of the associated atmospheric 5 deposition into aquatic and terrestrial ecosystems. Of the welfare effects categories listed in 6 section 302(h) of the Clean Air Act, the effects of oxides of nitrogen, oxides of sulfur and 7 particulate matter on aquatic and terrestrial ecosystems, which encompass soils, water, 8 vegetation, and wildlife, are the focus of this review. This PA focuses on the evidence described 9 in the 2020 ISA, and prior ISAs and AQCDs for the three criteria pollutants and focuses on 10 effects on specific ecosystems and biological receptors from N and S deposition and both the 11 confidence and key uncertainties associated with those effects. We also address considerations of the public welfare effects given that the public welfare implications of the evidence regarding S 12 13 and N related welfare effects are dependent on the type and severity of the effects, as well as the 14 extent of the effect at a particular biological or ecological level of organization. We discuss such 15 factors here in light of judgments and conclusions made in NAAQS reviews regarding effects on 16 the public welfare.

# 17 4.1 DIRECT EFFECTS OF OXIDES OF N AND S AND OF PM IN 18 AMBIENT AIR

19 There is a well-established body of scientific evidence that has shown that acute and 20 chronic exposures to oxides of N and S, such as SO<sub>2</sub>, NO<sub>2</sub>, NO, HNO<sub>3</sub> and PAN in the air, are 21 associated with negative effects on vegetation. Such scientific evidence, as was available in 22 1971, was the basis for the current secondary NAAQS for oxides of sulfur and oxides of 23 nitrogen, as summarized in section 3.1 above. The current scientific evidence continues to 24 demonstrate such effects, with the ISA specifically concluding that the evidence is sufficient to 25 infer a causal relationship between gas-phase SO<sub>2</sub> and injury to vegetation (ISA, Appendix 3, 26 section 3.6.12), and between gas-phase NO, NO<sub>2</sub> and PAN and injury to vegetation (ISA, 27 Appendix 3, section 3.6.2). The ISA additionally concluded the evidence to be sufficient to infer 28 a causal relationship between exposure to HNO<sub>3</sub> and changes to vegetation, noting that 29 experimental exposure can damage leaf cuticle of tree seedlings and HNO<sub>3</sub> concentrations have 30 been reported to have contributed to declines in lichen species in the Los Angeles basis (ISA, 31 Appendix 3, section 3.6.3). 32 Uptake of gas phase N and S pollutants in a plant canopy is a complex process involving

- adsorption to surfaces (leaves, stems and soil) and absorption into leaves (ISA, Appendix 3,
- 34 sections 3.1, 3.2 and 3.3). Several factors affect the extent to which ambient air concentrations of

1 gas-phase N and S pollutants elicit specific plant responses. These include rate of stomatal 2 conductance and plant detoxification mechanisms, and external factors such as plant water status, 3 light, temperature, humidity, and pollutant exposure regime (ISA Appendix 3, sections 3.2 and 4 3.3). The entry of gases into a leaf depends on atmospheric chemical processes and physical 5 characteristics of the surfaces, including the stomatal aperture. Stomatal opening is controlled 6 largely by environmental conditions, such as water availability, humidity, temperature, and light 7 intensity. When the stomata are closed, resistance to gas uptake is high and the plant has a very 8 low degree of susceptibility to injury (ISA, Appendix 3, section 3.1). However, "unlike vascular 9 plants, mosses and lichens do not have a protective cuticle barrier to gaseous pollutants, which is 10 a major reason for their sensitivity to gaseous S and N" (ISA, Appendix 3, p. 3-2).

11 Specifically for SOx, we note that high concentrations in the first half of the twentieth 12 century have been blamed for severe damage to plant foliage that occurred near large ore 13 smelters during that time (ISA, Appendix 3, section 3.2). In addition to foliar injury, which is 14 usually a rapid response, SO<sub>2</sub> exposures have also been documented to reduce plant 15 photosynthesis and growth. The appearance of foliar injury can vary significantly among species 16 and growth conditions (which affect stomatal conductance). The research on SO<sub>2</sub> effects on 17 vegetation has declined since the 1980s, especially in the U.S., due to the appreciable reductions 18 in ambient air concentrations of SO<sub>2</sub> (ISA, Appendix 3, section 3.2). For lichens, damage from 19 SO<sub>2</sub> exposure has been observed to include decreases in photosynthesis and respiration, damage 20 to the algal component of the lichen, leakage of electrolytes, inhibition of nitrogen fixation, 21 decreased potassium absorption, and structural changes (ISA, Appendix 3, section 3.2; Belnap et 22 al., 1993; Farmer et al., 1992, Hutchinson et al., 1996).

23 Although there is evidence of plant injury associated with SO<sub>2</sub> exposures dating back 24 more than a century (ISA, Appendix 3, section 3.2), as exposures have declined in the U.S., 25 some studies in the eastern U.S. have reported increased growth in some SO<sub>2</sub>-sensitive tree 26 species. For example, studies by Thomas et al. (2013) with eastern red cedar in West Virginia 27 have reported significant growth rate increases in more recent years. In this study, the authors 28 conducted a multivariate correlation analysis using historical climate variables, atmospheric CO<sub>2</sub> 29 concentrations, and estimated emissions of  $SO_2$  and  $NO_x$  in the U.S. found that the growth of 30 eastern red cedar trees (assessed through 100-year tree ring chronology) is explained best by 31 increases in atmospheric CO<sub>2</sub> and NO<sub>X</sub> emissions and decreases in SO<sub>2</sub> emissions. Although the 32 authors attributed the growth response to reductions in SO<sub>2</sub>-associated acid deposition, and 33 related recovery from soil acidification, the relative roles of different pathways is unclear as a 34 historical deposition record was not available (ISA, Appendix 3, section 3.2). Other researchers 35 have suggested that the observed red cedar response was related to the fact that the trees were 36 growing on a limestone outcrop that could be well buffered from soil acidification (Schaberg et

1 al., 2014). This seems to suggest a somewhat faster recovery than might be expected from 2 deposition-related soil acidification which may indicate a relatively greater role for changes in 3 ambient air concentrations of SO<sub>2</sub>, in combination with changes in other gases than was 4 previously understood (ISA, Appendix 3, section 3.2 and Appendix 5, section 5.2.1.3). 5 The evidence base evaluated in the 1993 AQCD for Oxides of N included evidence of 6 phytotoxic effects of NO, NO<sub>2</sub>, and PAN on plants through decreasing photosynthesis and 7 induction of visible foliar injury (U.S. EPA, 1993). The 1993 AQCD additionally concluded that 8 concentrations of NO, NO<sub>2</sub>, and PAN in the atmosphere were rarely high enough to have 9 phytotoxic effects on vegetation. Little new information is available since that time on these 10 phytotoxic effects at concentrations currently observed in the U.S. (ISA, Appendix 3, section 11 3.3). 12 The evidence for HNO<sub>3</sub> indicates a role in lichen species declines observed in the 1970s 13 in the Los Angeles basin (ISA, Appendix 3, section 3.3; Boonpragob and Nash 1991; Nash and

14 Sigal, 1999; Riddell et al., 2008). A 2008 resampling of areas shown to be impacted in the past 15 by HNO<sub>3</sub> found community shifts, declines in the most pollutant-sensitive lichen species, and 16 increases in abundance of nitrogen-tolerant lichen species compared to 1976–1977, indicating 17 that these lichen communities have not recovered and had experienced additional changes (ISA, Appendix 3, section 3.4; Riddell et al., 2011). The recently available evidence on this topic also 18 19 included a study of six lichen species that reported decreased chlorophyll content and 20 chlorophyll fluorescence, decreased photosynthesis and respiration, and increased electrolyte 21 leakage from HNO<sub>3</sub> exposures for 2-11 weeks (daily peak levels near 50 ppb) in controlled

chambers. (ISA, Appendix 3, section 3.4; Riddell et al., 2012).

23 Studies involving ambient air PM have generally involved conditions that would not be 24 expected to meet the current secondary standards for PM, e.g., polluted locations in India or 25 Argentina (ISA, Appendix 15, sections 15.4.3 and 15.4.4). Similarly, reduced photosynthesis has 26 been reported for rice plants experiencing fly ash particle deposition of 0.5 to  $1.5 \text{ g/m}^2$ -day, a 27 loading which corresponds to greater than 1000 kg/ha-yr (ISA, Appendix 15, sections 15.4.3 and 28 15.4.6). Further, studies of the direct effects of PM in ambient air on plant reproduction in near 29 roadway locations in the U.S. have not reported a relationship between PM concentrations and 30 pollen germination (ISA, Appendix 15, section 15.4.6). Rather, the evidence related to PM is that 31 associated with deposition of its components, as summarized in section 4.2.3 below.

32 4

# 4.2 DEPOSITION-RELATED ECOLOGICAL EFFECTS

As summarized in section 2.5 above, oxides of N and S, and PM, in ambient air
 contribute to deposition of N and S, which can affect ecosystem biogeochemistry, structure, and
 function in multiple ways. These effects include nutrient enrichment, primarily associated with
1 excess N, and acidification, due to N and S deposition. Both N and S are essential nutrients. 2 Nitrogen availability, however, is sometimes the limiting factor for plant growth and productivity 3 in aquatic and terrestrial ecosystems.<sup>1</sup> Accordingly, increases in the inputs of N-containing 4 compounds to an ecosystem can affect vegetation growth and productivity, which in natural 5 systems (both aquatic and terrestrial) can affect the relative representation and abundance of 6 different species as a result of differing N requirements and growth characteristics among 7 different species. Sulfur and N compounds can contribute to the acidity of terrestrial and aquatic 8 ecosystems. The extent to which S and N deposition contribute to ecosystem acidification or to 9 which N deposition contributes to nitrogen enrichment, and associated ecological effects, 10 depends on characteristics of the deposited compounds and the receiving ecosystem.

11 Ecosystem effects considered in the currently available evidence include effects on the 12 presence and abundance of different species, with the associated potential for changes in 13 ecosystem function (ISA, section IS.2.2.4). The ecological metrics that have commonly been 14 assessed, and for which there are effects related to atmospheric deposition, include species 15 richness, community composition and biodiversity. Species richness is the number of species in a 16 particular community and community composition additionally accounts for the number of 17 individuals of each species. For example, two sites may both have 10 species of trees but differ 18 in tree community composition because one may have nearly all individuals from one species 19 and the second may have equal representation by all 10 species. (ISA, section IS.2.2.4). 20 In addition to atmospheric deposition, other sources of S and N can play relatively greater 21 or lesser roles in contributing to S and N inputs, depending on location. For example, many 22 waterbodies receive appreciable amounts of N from agricultural runoff and municipal or 23 industrial wastewater discharges. Additionally, the impacts of historic deposition in both aquatic

and terrestrial ecosystems pose complications to discerning the potential effects of more recentlower deposition rates.

Another complication specific to N deposition is its potential to increase growth and yield of agricultural and timber crops, which may be judged and valued differentially than changes in growth of some species in natural ecosystems (as noted in section 4.3 below). As discussed further in section 4.2.2 below, N enrichment in natural ecosystems can, by increasing growth of N limited plant species, change competitive advantages of species in a community, with associated impacts on the composition of the ecosystem's plant community. The following sections draw from the ISA to provide an overview of the welfare effects

33 associated with N and S deposition ecosystems of the U.S. Section 4.2.1 focuses on acidification-

<sup>&</sup>lt;sup>1</sup> In addition to N, phosphorus is the other essential nutrient for which availability sometimes is the limiting factor in plant growth and productivity, e.g., in many aquatic systems. Sulfur is rarely limiting in natural systems (ISA, Appendix 7, section 7.1 and Appendix 4, section 4.3).

related effects, while 4.2.2 focuses on effects related to nitrogen enrichment. Lastly, section 4.3.2
provides an overview of other deposition-related effects. The summaries in the sections and their
subsections below are organized in a manner intended to address the following questions.

- What is the nature of the welfare effects associated with N and S and PM deposition? Is there new evidence on welfare effects beyond those identified in the last review? Does the newly available evidence alter prior conclusions?
- What does the available evidence indicate regarding ecosystems at particular risk from deposition-related effects, and what are associated important, or key, uncertainties?
- What are important uncertainties in the evidence? To what extent have such
   uncertainties identified in the evidence in the past been reduced and/or have new
   uncertainties been recognized?
- 13 4.2.1 Acidification and Associated Effects

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14 Deposited S and N compounds can both act as acidifying agents. Acidifying deposition 15 can affect biogeochemical processes in soils, with ramifications for terrestrial biota and for the 16 chemistry and biological functioning of associated surface waters (ISA, Appendix 7, section 7.1). 17 Soil acidification is influenced by the deposition of inorganic acids (HNO<sub>3</sub> and  $H_2SO_4$ ), and by 18 chemical and biological processes, which can also be influenced by atmospheric deposition of 19 other chemicals. For example,  $NH_3$  or  $NH_4^+$  can stimulate soil bacteria that produce  $NO_3^-$  (ISA, 20 Appendix 4, section 4.3). In this process, a hydrogen ion is produced and the extent to which this changes soil acidity depends on the fate of the NO<sub>3</sub><sup>-</sup>. When NO<sub>3</sub><sup>-</sup>, or SO<sub>4</sub><sup>2-</sup>, leach from soils to 21 22 surface waters, an equivalent amount of positive cations, or countercharge, is also transported. If the countercharge is provided by a base cation (e.g., calcium,  $[Ca^{2+}]$ , magnesium  $[Mg^{2+}]$ , sodium 23  $[Na^+]$ , or potassium  $[K^+]$ ), rather than hydrogen  $(H^+)$ , the leachate is neutralized, but the soil 24 25 becomes more acidic from the H<sup>+</sup> left behind and the base saturation of the soil is reduced by the loss of the base cation. Depending on the relative rates of soil processes that contribute to the soil 26 pools of H<sup>+</sup> and base cations, such as weathering, continued SO<sub>4</sub><sup>2-</sup> or NO<sub>3</sub><sup>-</sup> leaching can deplete 27 28 the soil base cation pool which contributes to increased acidity of the leaching soil water, and by 29 connection, the surface water. Accordingly, the ability of a watershed to neutralize acidic 30 deposition is determined by a variety of biogeophysical factors including weathering rates, 31 bedrock composition, vegetation and microbial processes, physical and chemical characteristics 32 of soils, and hydrology (ISA Appendix 4, section 4.3). 33 This connection between  $SO_2$  and  $NO_x$  emissions, atmospheric deposition of N and/or S,

- 34 and the acidification of acid-sensitive soils and surface waters is well documented with several
- decades of evidence, particularly in the eastern U.S. (ISA, section IS.5; Appendix 8, section 8.1).
- 36 While there is evidence newly available since the 2008 ISA, in general, the fundamental

understanding of mechanisms and biological effects has not changed. Rather, the more recent
 studies further support the 2008 ISA findings on these broad conclusions and provide updated
 information on specific aspects. An overview of the ISA findings is provided for aquatic
 acidification in section 4.2.2 below, and for terrestrial acidification in section 4.2.3 below.

5

#### 4.2.1.1 Freshwater Ecosystems

6 Surface water processes integrate the chemicals deposited directly onto waterbodies with 7 those released from hydrologically connected terrestrial ecosystems as a result of deposition 8 within the watershed (ISA, Appendix 7, section 7.1). As was the case in the last review, the body 9 of evidence regarding such processes available in this review, including that newly available, is 10 sufficient to infer a causal relationship between N and S deposition and the alteration of 11 freshwater biogeochemistry (ISA, section IS.6.1). Additionally, based on the previously 12 available evidence, the current body of evidence is also sufficient to conclude that a causal 13 relationship exists between acidifying deposition and changes in biota, including physiological 14 impairment and alteration of species richness, community composition, and biodiversity in 15 freshwater ecosystems (ISA, section IS.6.3).

16 In addition to the acidity of surface waters quantified over weeks or months, waterbodies 17 can also experience spikes in acidity in response to episodic events such as precipitation or rapid 18 snowmelt that may elicit a pulse of acidic leachate over shorter periods such as hours or days. In 19 these situations, sulfate and nitrate in snowpack (or downpours) can provide a surge or pulse of 20 drainage water, containing acidic compounds, that is routed through upper soil horizons rather 21 than the deeper soil horizons that usually would provide buffering for acidic compounds (ISA, 22 Appendix 7, section 7.1). During these episodes, N and S sources other than atmospheric 23 deposition, such as acid mine drainage or road salt applications can also be important. While 24 some streams and lakes may have chronic or base flow chemistry that provides suitable 25 conditions for aquatic biota, they may experience occasional acidic episodes with the potential 26 for deleterious consequences to sensitive biota (ISA, Appendix 8, section 8.5).

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#### 4.2.1.1.1 Nature of Effects and New Evidence

Longstanding evidence has well characterized the changes in biogeochemical processes and water chemistry caused by N and S deposition to surface waters and their watersheds and the ramifications for biological functioning of freshwater ecosystems (ISA, Appendix 8, section 8.1).

- 31 The 2020 ISA found that the newly available scientific research "reflects incremental
- 32 improvements in scientific knowledge of aquatic biological effects and indicators of acidification
- as compared with knowledge summarized in the 2008 ISA" (ISA, Appendix 8, p. 8-80).
- 34 Previously and newly available studies "indicate that aquatic organisms in sensitive ecosystems
- 35 have been affected by acidification at virtually all trophic levels and that these responses have

1 been well characterized for several decades" (ISA, Appendix 8, p. 8-80). For example,

- 2 information reported in the previous 2008 ISA "showed consistent and coherent evidence for
- 3 effects on aquatic biota, especially algae, benthic invertebrates, and fish that are most clearly
- 4 linked to chemical indicators of acidification" (ISA, Appendix 8, p. 8-80). These indicators are
- 5 surface water pH, base cation ratios, acid neutralizing capacity (ANC), and inorganic aluminum
- 6 (Al<sub>i</sub>) concentration (ISA, Appendix 8, Table 8-9).

7 The effects of aquatic acidification on fish species are especially well understood in the 8 scientific literature, and many species have been documented to have experienced negative 9 effects from acidification (ISA, Appendix 8, section 8.3). Research conducted in fresh 10 waterbodies of Europe and North America before 1990 documented the adverse biological 11 effects on various fish species associated with acidification (ISA, Appendix 8, section 8.3.6). 12 Some of the most commonly studied fish species are brown and brook trout, and Atlantic 13 salmon, among these species the earliest lifestages are most sensitive to acidic conditions. Many 14 effects of acidic surface waters on fish, particularly effects on gill function or structure, relate to 15 the combination of low pH and elevated inorganic Al (ISA, Appendix 8, section 8.3.6.1).

Based on studies in the 1980s and 1990s of waterbodies affected by acidic deposition, researchers have summarized the evidence of effects on fish populations in relation to the pH and ANC of the studied waterbodies. Such effects include reduced presence of some species in acidified lakes in the Adirondacks of New York or the Appalachian Mountains (ISA, Appendix 8, section 8.3.6). Such studies have been used to characterize ranges of ANC as to potential risk to aquatic communities. The use of ANC as an indicator of waterbody acidification is described in section 4.2.1.1.2 below.

23 Despite the reductions in acidifying deposition, as summarized in section 2.5 above, 24 aquatic ecosystems across the U.S. are still experiencing effects from historical contributions of 25 N and S (ISA, Appendix 8, section 8.6). Long-term monitoring programs in several acid-26 sensitive regions of the U.S., including the Adirondacks and the northeastern U.S. have 27 documented temporal trends in surface water chemistry that include evidence for chemical 28 recovery in the northeastern and southeastern U.S. suggesting that full chemical recovery may 29 take many decades or not occur at all due to the dynamics of S adsorption and desorption and 30 long-term Ca depletion of soils (ISA, Appendix 7, section 7.1.5.1, Appendix 11, section 11.2 and 31 Appendix 16, section 16.3.4). As reported in the 2008 ISA, biological recovery of aquatic 32 systems lags chemical recovery due to a number of physical and ecological factors (including the 33 time for populations to recover), as well as other environmental stressors, which make the time 34 required for biological recovery uncertain (ISA, section 8.4). Some recent studies report on 35 waterbodies showing signs of recovery from the impacts of many decades of substantially 36 elevated acidic deposition. One example is the successful reintroduction and re-establishment of

1 a naturalized native fish species (brook trout) in an Adirondack Lake from which the species had

- 2 been previously lost. Based on reconstruction of the historical record, the study reported ANC
- 3 had increased from -2  $\mu$ eq/L during the 1980s to 12  $\mu$ eq/L during the period 2010-2012 when the
- 4 trout were reintroduced. By 2012, young fish were observed, documenting successful
- 5 reproduction in or in tributary streams near, the lake (ISA, Appendix 8, section 8.4.4; Sutherland
- 6 et al., 2015). Another recent study in the Adirondack Lake region however, found no evidence of
- 7 widespread or substantial brook trout recovery, although water quality had improved, indicating
- 8 the impact of the factors mentioned above that can contribute to lags of biological recovery
- 9 behind chemical recovery (ISA, Appendix 8, sections 8.4 and 8.4.4).
- 10

# 4.2.1.1.2 Freshwater Ecosystem Sensitivity

11 The effects of acid deposition on aquatic systems depend largely upon the ability of the 12 system to neutralize additional acidic inputs from the environment, whether from the atmosphere 13 or from surface inputs. There is a large amount of variability between freshwater systems in this 14 regard which reflects their underlying geology as well as previous acidic inputs. Accordingly, 15 different freshwater systems (e.g., in different geographic regions) respond differently to similar 16 amounts of acid deposition. The main factor in determining sensitivity is the underlying geology 17 of an area and its ability to provide soil base cations through weathering to buffer acidic inputs 18 (ISA, Appendix 8, section 8.5.1). As noted in the ISA, "[g]eologic formations having low base 19 cation supply, due mainly to low soil and bedrock weathering, generally underlie the watersheds 20 of acid-sensitive lakes and streams" (ISA, Appendix 8, p. 8-58). Consistent with this, studies 21 have indicated that the thickness of the till (the sediment layer deposited by action of receding 22 glaciers) "has been shown to be a key control on the pH and ANC of Adirondack lakes" (ISA, 23 Appendix 8, p. 8-58). Other factors identified as contributing to the sensitivity of surface waters 24 to acidifying deposition, include topography, soil chemistry and physical properties, land use and 25 history, and hydrologic flowpath, as well as impacts of historic, appreciably higher, deposition 26 (ISA, Appendix 8, p. 8-58).

Acid neutralizing capacity (ANC) is commonly used to describe the potential sensitivity of a freshwater system to acidification-related effects and has been found in various studies to be the single best indicator of the biological response and health of aquatic communities in acid sensitive systems (ISA, Appendix 8, section 8.6). This indicator is defined as the molar sum of strong base cations minus the molar sum of strong acid anions:

32

ANC = 
$$(Ca^{2+} + Mg^{2+} + K^{+} + Na^{+} + NH_{4}^{+}) - (SO_{4}^{2-} + NO_{3}^{-} + Cl^{-})$$

33 While ANC is not the direct cause of acidification-related effects on aquatic biota, it serves as an

34 indicator of acidification-related risk. Water quality models are generally better at estimating

35 ANC than other indicators and ANC has been related to the health of biota and other surface

1 water constituents like pH and Al or watershed components like base cation weathering (BCw)

- 2 (ISA, Appendix 8, sections 8.1 and 8.3.6.3). Waterbody pH largely controls the bioavailability of
  3 Al, which is very toxic to fish (ISA, Appendix 8, section 8.6.4).
- 4 In its role as an indicator, ANC levels are commonly used to categorize waterbody 5 sensitivity. Waterbodies with annual average levels above 100 are generally not considered 6 sensitive or at risk of acidification-related effects. There is potential for risk at lower levels, at 7 which consideration of other factors can inform interpretation. National survey data dating back 8 to the early 1980s that were available for the 2008 ISA indicated acidifying deposition had 9 acidified surface waters in the southwestern Adirondacks, New England uplands, eastern portion 10 of the upper Midwest, forested Mid-Atlantic highlands, and Mid-Atlantic coastal plain (2008 11 ISA, section 4.2.2.3; ISA, Appendix 8, section 8.5.1). As noted in section 4.2.1.1 above, events 12 such as spring snowmelt and heavy rain events can contribute to episodic acidification events. 13 For example, in some impacted northeastern waterbodies, ANC levels may dip below zero for 14 hours to days or weeks in response to such events, while waterbodies labeled chronically acidic 15 have ANC levels below zero throughout the year (ISA, section IS6.1.1.1; Driscoll et al 2001).
- 16 Accordingly, headwater streams in both the eastern and western U.S. tend to be more sensitive to
- 17 such episodes due to their smaller size (ISA, Appendix 8, section 8.5.1).
- 18 Fish and water quality surveys as well as *in situ* bioassays inform our understanding of 19 risk posed to fish species across a range of ANC. For example, surveys in the heavily impacted 20 Adirondack mountains found that waterbodies with ANC levels near/below zero<sup>2</sup> and pH 21 near/below 5.0 generally had few or no fish species (Sullivan et al., 2006a; ISA, Appendix 8, 22 section 8.6). Waterbodies with levels of ANC above zero differed in the types and numbers of 23 species present. At relatively lower ANC levels such as below 20 µeq/L, comparatively acid 24 tolerant species such as brook trout can have healthy populations, but sensitive fish species such 25 as Atlantic salmon smolts, blacknose shiner, and other fish can be absent, or their population can 26 be greatly reduced. While most sensitive species were not lost from the aquatic system, their 27 fitness (population size and growth) declined; plankton and macroinvertebrate assemblages were 28 also impacted somewhat; and fish species richness in some areas was lower with fewer of the 29 most sensitive species. Some sites with ANC levels above 80 µeq/L have appeared unimpaired 30 (Bulger et al., 1999; Driscoll et al., 2001; Kretser et al., 1989; Sullivan et al., 2006). An ANC 31 level of 100 µeq/L is often identified as a benchmark at/below which waterbodies may be
- 32 considered at increased sensitivity.

 $<sup>^{2}</sup>$  A survey of waterbodies in the Adirondacks in 1984-1987 found 27% of streams to have ANC values below zero, with a minimum value of -134  $\mu$ eq /L (Sullivan et al., 2006). Values of ANC below 20 in Shenandoah stream sites were associated with fewer fish of sensitive species compared to sites with higher ANC (Bulger et al., 1999).

- 1 Surveys conducted from the 1980s through 2004, available in the last review, indicated 2 that the surface waters in the southwestern Adirondacks, New England uplands, eastern portion 3 of the upper Midwest, forested Mid-Atlantic highlands, and Mid-Atlantic coastal plain had been 4 acidified as a result of acidifying deposition (ISA, Appendix 8, section 8.5.1). A compilation of 5 historical water quality measurements of ANC from 1980 to 2011 (nearly 200,000 measurements
- 6 at nearly 20,000 spatially unique sites) is presented in Figure 4-1 below (Sullivan, 2017).<sup>3</sup> As
- 7 described in the ISA, "[a]cidic waters were mostly restricted to northern New York, New
- 8 England, the Appalachian Mountain chain, upper Midwest, and Florida" (ISA, Appendix 8, p. 8-
- 9 60). Additionally, the figure indicates low, but positive, ANC values for these same regions, as
- 10 well as high-elevation western waterbodies (e.g., in the Sierra and Cascades mountains) and parts
- 11 of Arkansas and the Gulf states (Figure 4-1; ISA, Appendix 8, section 8.5.2). The findings for
- 12 high-elevation portions of the West and parts of Arkansas and the Gulf states are thought to
- 13 largely reflect base cation supply in soils, as levels of acidifying deposition have been low in
- 14 most areas of the West, and acidic surface waters there are rare (ISA, section 8.5.2).

<sup>&</sup>lt;sup>3</sup> Samples expected to be strongly influenced by acid mine drainage, sea salt spray, or road salt application were excluded. Among the full dataset, 6,065 sites had ANC < 100  $\mu$ eq/L.



Figure 4-1. Surface water ANC map, based on data compiled by Sullivan (2017) (ISA, Appendix 8, Figure 8-11).

#### 1

#### 4.2.1.1.3 Key Uncertainties

2 In the longstanding evidence base for acidification effects of deposited S and N in aquatic 3 ecosystems, uncertainties remain. Key uncertainties include those associated with inputs to 4 models that simulate watershed chemistry and are employed to estimate waterbody buffering 5 capacity including weathering rates and leaching. Uncertainties are associated with estimates of 6 the response of waterbodies to different deposition levels in areas for which site-specific data are 7 not available because of the high spatial variability of the factors that influence watershed 8 sensitivity (ISA, Appendix 8, section 8.5.1; McNulty et al., 2007). For example, there are 9 uncertainties related to limitations in water quality measurements, data on surface runoff 10 characteristics, and other factors important to characterizing watershed supplies of base cations 11 related to weathering of bedrock and soils. There are also uncertainties associated with our 12 understanding of relationships between ANC and risk to native biota, particularly in waterbodies 13 in geologic regions prone to waterbody acidity. These relate to the varying influences of site-14 specific factors other than ANC.

15

#### 4.2.1.2 Terrestrial Ecosystems

16 There is longstanding evidence that changes in soil biogeochemical processes caused by 17 acidifying deposition of N and S to terrestrial systems are linked to changes in terrestrial biota, 18 with associated impacts on ecosystem characteristics. The currently available evidence, including 19 that newly available in this review, supports and strengthens this understanding (ISA, Appendix 20 5, section 5.1). Consistent with conclusions in the last review the current body of evidence is 21 sufficient to infer a causal relationship between acidifying deposition and alterations of 22 biogeochemistry in terrestrial ecosystems. Additionally, and consistent with conclusions in the 23 last review, the current body of evidence is sufficient to infer a causal relationship between 24 acidifying N and S deposition and the alteration of the physiology and growth of terrestrial 25 organisms and the productivity of terrestrial ecosystems. The current body of evidence is also 26 sufficient to conclude that a causal relationship exists between acidifying N and S deposition and 27 alterations of species richness, community composition, and biodiversity in terrestrial 28 ecosystems (2008 ISA, sections 4.2.1.1 and 4.2.1.2; 2020 ISA, sections 4.1, 5.7.1 and 5.7.2).

29

#### 4.2.1.2.1 Nature of Effects and New Evidence

30 Deposition of acidifying compounds to acid-sensitive soils can cause soil acidification, 31 increased mobilization of Al from soil to drainage water, and deplete the pool of exchangeable 32 base cations in the soil (ISA, Appendix 5, section 5.2 and Appendix 4, sections 4.3.4 and 4.3.5). 33 The physiological effects of acidification on terrestrial biota include slower growth and increased 34 mortality among sensitive plant species, which are generally attributable to physiological 35 impairment caused by Al toxicity (related to increased availability of inorganic Al in soil water)

1 and a reduced ability of plant roots to take up base cations (ISA, Appendix 4, section 4.3 and 2 Appendix 5, section 5.2). The U.S. tree species most studied with regard to effects of acid 3 deposition are red spruce and sugar maple, although there is also evidence for other tree species 4 such as flowering dogwood (ISA, Appendix 5, section 5.2.1). The recently available evidence 5 includes Ca addition experiments in which Ca is added to acidic soils and physiological and 6 growth responses of red spruce and sugar maple are assessed to help understand the response of 7 these species to the soil changes induced by acid deposition (ISA, Appendix 5, Table 5-2). Other 8 recent studies have included addition or gradient studies evaluating relationships between soil 9 chemistry indicators of acidification (e.g., soil pH, Bc:Al ratio, base saturation and Al) and 10 ecosystem biological endpoints, including physiological and community responses of trees and 11 other vegetation, lichens, soil biota, and fauna (ISA, Appendix 5, Table 5-6). 12 Since the last review of the NAAQS for oxides of S and N, and as described in detail in 13 Chapter 5 (and Appendix 5B), several observational studies have reported on statistical 14 associations between tree growth or survival, as assessed at monitoring sites across the U.S. and 15 estimates of average deposition of S or N compounds at those sites over time periods on the 16 order of 10 years (section 5.4.2.3 and Appendix 5B, section 5B.2.2 below; ISA, Appendix 5, 17 section 5.5.2 and Appendix 6, section.6.2.3.1; Dietze and Moorcroft, 2011; Thomas et al., 2010; 18 Horn et al., 2018). Negative associations were observed for survival and growth in a number of 19 species or species groups with S deposition metrics; positive and negative associates were 20 reported with N deposition (see section 5.4.2.3 and 5.5.3 below and Appendix 5B). 21 The physiological effects of acidifying deposition on terrestrial biota can also result in 22 changes in species composition whereby sensitive species are replaced by more tolerant species, 23 or the sensitive species that were dominant in the community become a minority. For example, 24 increasing soil cation availability (as in Ca addition or gradient experiments) was associated with 25 greater growth and seedling colonization for sugar maple while American beech was more 26 prevalent on soils with lower levels of base cations where sugar maple does less well (ISA, 27 Appendix 5, section 5.2.1.3.1; Duchesne and Ouimet, 2009). In a study of understory species 28 composition, soil acid-base chemistry was found to be a predictor of understory species 29 composition (ISA, Appendix 5, section 5.2.2.1). Additionally, limited evidence, including a 30 recent S addition study and agricultural soil gradient study, indicated that soil acid-base 31 chemistry predicted and was correlated with diversity and composition of soil bacteria, fungi, 32 and nematodes (ISA, Appendix 5, section 5.2.4.1). 33 4.2.1.2.2 Terrestrial Ecosystem Sensitivity 34 Underlying geology is the principal factor governing the sensitivity of both terrestrial and 35 aquatic ecosystems to acidification from S and N deposition. Geologic formations with low base

1 cation supply (e.g., sandstone, quartzite), due mainly to low weathering, generally underlie these

- 2 acid sensitive watersheds. Other factors also contribute to the overall sensitivity of an area to
- 3 acidifying nitrogen and sulfur deposition including topography, soil chemistry, land use, and
- 4 hydrology (ISA, Appendix 5, section 5.3). As observed in the ISA, "[a]cid-sensitive ecosystems
- 5 are mostly located in upland mountainous terrain in the eastern and western U.S. and are
- 6 underlain by bedrock that is resistant to weathering, such as granite or quartzite sandstone" (ISA,
- 7 Appendix 7, p. 7-45). Further, as documented in the evidence, biogeochemical sensitivity to
- 8 deposition-driven acidification (and eutrophication [see section 4.2.2 below]) is the result of
- 9 historical loading, geologic/soil conditions (e.g., mineral weathering and S adsorption), and
- 10 nonanthropogenic sources of N and S loading to the system (ISA, Appendix 7, section 7.1.5).
- 11 Several different indicators are commonly used to identify areas at increased risk of 12 acidification processes (ISA, Appendix 5, Table 4-1). They include the ratio of the molar sum of 13 base cations to the molar amount of Al (BC:Al). The BC:Al ratio is commonly used, particularly in mass balance modeling approaches, such as the simple mass balance equation (SMB), that are 14 15 intended to assess the vulnerability of different areas to acidification as a result of atmospheric 16 deposition of N and S compounds. Higher values of this ratio indicate a lower potential for 17 acidification-related biological effects (ISA, Table IS-2). The ratio value can be reduced by 18 release of base cations from the soil (e.g., through the process of neutralizing drainage water 19 acidity) which, in turn, reduces the base saturation of the soil. Soil base saturation<sup>4</sup> and changes 20 to it can also be an indicator of acidification risk (ISA, Appendix 4, section 4.3.4). The 21 accelerated loss of base cations through leaching, decrease in base saturation, and decreases in 22 the BC:Al ratio all serve as indicators of soil acidification. This is because the input of base 23 cations to soil solution, e.g., via soil weathering or base cation exchange, can neutralize
- 24 inorganic and organic acids (ISA, Appendix 4, section 4.3).
- 25 Although there has been no systematic national survey of U.S. terrestrial ecosystem soils, 26 several forest ecosystems are considered the most sensitive to terrestrial acidification from 27 atmospheric deposition. These include forests of the Adirondack Mountains of New York, Green 28 Mountains of Vermont, White Mountains of New Hampshire, the Allegheny Plateau of 29 Pennsylvania, and mountain top and ridge forest ecosystems in the southern Appalachians (2008) 30 ISA, Appendix 3, section 3.2.4.2; ISA, Appendix 5, section 5.3). A number of modeling 31 analyses, including a national-scale analysis, have been performed to identify acid-sensitive 32 areas, generally through estimates of indicators such as BC:Al (ISA, Appendix 5, sections 5.3,
- 33 5.4 and 5.5). In some cases, more recent analyses augment estimates from the previously

<sup>&</sup>lt;sup>4</sup> Soil base saturation expresses the concentration of exchangeable bases (Ca, Mg, potassium [K], sodium [Na]) as a percentage of the total cation exchange capacity (which includes exchangeable H+ and inorganic Al).

1 available national-scale analysis (McNulty et al., 2007), potentially providing updated estimates.

- 2 For example, a recent modeling analysis by Phelan et al. (2014) employed the PROFILE model
- 3 to estimate base cation weathering (BC<sub>W</sub>) in support of simple mass balance (SMB) modeling, a
- 4 difference from the empirical approach (clay correlation-substrate method) used by McNulty et
- 5 al.,2007. This more recent analysis suggested that Pennsylvania hardwood sites may not be as
- 6 sensitive to acidifying deposition as previously estimated (ISA, Appendix 5, section 5.4; Phelan
- 7 et al., 2014). Another commonly used indicator of acidification is soil base saturation (ISA,
- 8 Appendix 4, Table 4-1). Values below 10% have been associated with areas experiencing
- 9 acidification such as the eastern forests recognized above (ISA, Appendix 4, section, 4.3.4).
- 10 Recently available evidence includes some studies describing early stages of recovery 11 from soil acidification in some eastern forests. For example, studies at the Hubbard Brook 12 Experimental Forest in New Hampshire reported indications of acidification recovery in soil 13 solution measurements across the period from 1984 to 2011 (ISA, Appendix 4, section 4.6.1; 14 Fuss et al., 2015). Another study of 27 sites in eastern Canada and the northeastern U.S. reported reductions in wet SO<sub>4</sub><sup>2-</sup> deposition to be positively correlated with changes in base saturation and 15 16 negatively correlated with changes in exchangeable Al between initial samplings in the mid 17 1980s to early 1990s and a resampling in the period 2003-2014. That is, reductions in wet deposition SO<sub>4</sub><sup>2-</sup> were associated with increases in soil base saturation and decreases in 18 19 exchangeable Al (ISA, Appendix 4, section 4.6.1; Lawrence et al., 2015). Modeling analyses 20 indicate extended timeframes for recovery are likely, as well as delays or lags related to 21 accumulated pools of S in forest soils (ISA, Appendix 4, section 4.6.1).
- 22

# 4.2.1.2.3 Key Uncertainties

23 Although the evidence clearly demonstrates that acidifying deposition of N and S causes 24 acidification related effects in terrestrial ecosystems, uncertainties remain that are important to 25 our consideration of the evidence in this review. For example, there are uncertainties associated 26 with the various approaches for estimating sensitive ecosystems and for understanding and 27 characterizing long-term risks and processes against the backdrop of deposition reductions 28 occurring over the past several decades. As summarized in section 4.2.1.2.2 above, modeling 29 analyses are commonly employed, with several inputs recognized as contributing to overall 30 uncertainty.

As noted in the ISA, the rate of base cation weathering "is one of the most influential yet difficult to estimate parameters" in modeling (such as the SMB) that estimate indicators of acidification as a function of deposition inputs (ISA, Appendix 4, section 4.5.1.1). Estimating this parameter continues to be a major source of uncertainty in such modeling. For example, in an analysis of uncertainties associated with simulating ANC in waterbodies of interest in

1 response to acid deposition over a broad spatial scale, the primary source of uncertainty was 2 identified to be from components of BCw (Li and McNulty, 2007). The authors concluded that 3 improvements in estimates of these components are crucial to reducing uncertainty and 4 successful model application for broader scales (e.g., where site-specific information is limited), 5 including national scale (ISA, Appendix 4, section 4.6). Another analysis of major sources of 6 uncertainty related to estimating soil acidification also found the greatest uncertainty to be 7 associated with the BCw estimates, particularly citing the particle size class-based method 8 commonly used to estimate the total specific surface area upon which weathering reactions can 9 take place (Whitfield et al., 2018).

10 There are also more general sources of uncertainty associated with observational or 11 gradient studies that relate variation in biological/ecological indices to variation in deposition 12 metrics. For example, studies may fail to account for influences such as variation in biological 13 and biogeochemical processes imposed by climate, geology, biota, and other environmental 14 factors. Further, observed variation in current or recent biological metrics may be affected by the 15 lags reported in the evidence, both in ecosystem response to acid deposition and to ecosystem 16 recovery from historic deposition. Additionally, biological measures in areas for which recent 17 deposition metrics are relatively low, may be influenced by impacts from past deposition.

#### 18 4.2.2 Nitrogen Enrichment and Associated Effects

19 The numerous ecosystem types that occur across the U.S. have a broad range of 20 sensitivity to N enrichment. Organisms in their natural environments are commonly adapted to 21 the nutrient availability in those environments. Historically, N has been the primary limiting 22 nutrient in many terrestrial and aquatic ecosystems. Because of this, most species are adapted to 23 low nutrient conditions, and a much smaller fraction of species are adapted to high nutrient 24 availability. Therefore, when limiting nutrients become more available, whether from 25 atmospheric deposition, runoff, or episodic events, often selection leads to a shift in the 26 community from high diversity systems to low diversity systems. Thus, change in the availability 27 of an important nutrient, such as nitrogen, can, in nitrogen-limited systems, affect growth and 28 productivity, with ramifications on relative abundance of different species, and potentially 29 further and broader ramifications on ecosystem processes, structure, and function. The term, 30 eutrophication, refers to such processes that occur in response to enrichment of a system with 31 nutrients. A common example of eutrophication in aquatic ecosystems occurs when increased 32 loading of the limiting nutrient (usually N or phosphorous) results in rapid and appreciable algal 33 growth. Decomposition of the plant biomass from the subsequent algal die-off contributes to 34 reduced waterbody oxygen which in turn contributes to fish mortality (ISA, p. ES-18).

Both N oxides and reduced forms of nitrogen (NH<sub>x</sub>) can contribute to N enrichment. For
 many terrestrial and freshwater ecosystems, sources of N other than atmospheric deposition,
 including fertilizer and waste treatment, contribute to ecosystem total N with contributions that
 can be larger than that from atmospheric deposition (ISA Appendix 7, sections 7.1 and 7.2).

5

#### 4.2.2.1 Aquatic and Wetland Ecosystems

6 Nitrogen additions, including from atmospheric deposition, to freshwater, estuarine and 7 near-coastal ecosystems can contribution to eutrophication which typically begins with nutrient-8 stimulated rapid algal growth developing into an algal bloom that can, depending on various site-9 specific factors, be followed by anoxic conditions associated with the algal die-off. This 10 reduction in dissolved oxygen can affect higher-trophic-level species (ISA, section ES.5.2). The 11 extensive body of evidence in this area is sufficient to infer causal relationships between N 12 deposition and the alteration of biogeochemistry in freshwater, estuarine and near-coastal marine 13 systems (ISA, Appendix, sections 7.1 and 7.2). Further, consistent with findings in the last 14 review, the current body of evidence is sufficient to infer a causal relationship between N 15 deposition and changes in biota, including altered growth and productivity, species richness, 16 community composition, and biodiversity due to N enrichment in freshwater ecosystems (ISA, 17 Appendix 9, section 9.1). The body of evidence is sufficient to infer a causal relationship 18 between N deposition and changes in biota, including altered growth, total primary production, 19 total algal community biomass, species richness, community composition, and biodiversity due 20 to N enrichment in estuarine environments (ISA, Appendix 10, section 10.1). 21 The impact of N additions on wetlands depends on the type of wetland and other factors. 22 More specifically, the type of wetland, as well as hydrological conditions and season, influence 23 whether a wetland serves as a source, sink, or transformer of atmospherically deposited N (ISA, 24 section IS.8.1 and Appendix 11, section 11.1). One of the transformations that may occur in 25 wetlands is denitrification which leads to the production of N<sub>2</sub>O, a greenhouse gas. This is a 26 normal process in anaerobic soils but can be increased with the introduction of additional N, 27 especially in reduced forms such as  $NH_4^+$  (ISA, section 4.3.6). Whether wetlands are a source 28 and/or a sink of N is extremely variable and depends on vegetation type, physiography, and local 29 hydrology, as well as climate. Studies generally show N enrichment to decrease the ability of 30 wetlands to retain and store N, which may diminish the wetland ecosystem service of improving 31 water quality (ISA, section IS.8.1). Consistent with the evidence available in the last review, the

32 current body of evidence is sufficient to infer a causal relationship between N deposition and the

33 alteration of biogeochemical cycling in wetlands. Newly available evidence regarding N inputs

- 34 and plant physiology, expands the evidence base related to species diversity. The currently
- 35 available evidence, including that newly available, is sufficient to infer a causal relationship

1 between N deposition and the alteration of growth and productivity, species physiology, species

- 2 richness, community composition, and biodiversity in wetlands (ISA, Appendix 11, section
- 3 11.10).
- 4

# 4.2.2.1.1 Nature of Effects and New Evidence

- 5 As summarized above, N inputs and other factors contribute to nutrient enrichment which
- 6 contribute to eutrophication, the process of enriching a water body with nutrients resulting in
- 7 increased growth and change in the composition of primary producers (algae and/or aquatic
- 8 plants) which can also lead to low oxygen levels in the water body when these primary producers
- 9 decompose. Such nitrogen driven eutrophication alters freshwater biogeochemistry and can
- 10 impact physiology, survival, and biodiversity of sensitive aquatic biota (Figure 4-2).



11

# Figure 4-2. Conceptual model of the influence of atmospheric N deposition on freshwater nutrient enrichment (ISA, Appendix 9, Figure 9-1).

14 Evidence newly available in this review provides insights regarding N enrichment and its

- 15 impacts in several types of aquatic systems, including freshwater streams and lakes, estuarine
- 16 and near-coastal systems, and wetlands. For example, studies published since the 2008 ISA
- 17 augment the evidence base for high-elevation waterbodies where the main source of N is
- 18 atmospheric deposition, including a finding that N deposition is correlated with a shift from N to
- 19 P limitation in certain water bodies (ISA, section 9.1.1.3). The newly available evidence,
- 20 including that from paleolimnological surveys, fertilization experiments, and gradient studies
- 21 continues to show effects of N loading to sensitive freshwater systems, including an influence on

1 the occurrence of harmful algal blooms (ISA, Appendix 9). More specifically, the availability

- 2 and form of N has been found to influence algal bloom composition and toxicity (ISA, Appendix
- 3 9, section 9.2.6.1). Such evidence is also available in estuarine systems. For example, specific

4 phytoplankton functional groups prefer reduced forms of N (such as  $NH_4^+$ ) over oxidized forms

5 (such as NO<sub>3</sub><sup>-</sup>), and in many parts of the U.S., including the Southeast and Mid-Atlantic, reduced

6 N deposition has increased relative to oxidized N deposition (ISA, Appendix 10, section 10.3.3).

7 Very limited evidence suggests a role for atmospheric N deposition in taxonomic shifts and

8 declines in some invertebrates, although "the effects attributed to N are difficult to separate from

9 other stressors such as climate change and invasive species" (ISA, Appendix 9, section 9.6).

Evidence in coastal waters has recognized a role of nutrient enrichment in acidification of some coastal waters (ISA, Appendix 10, section 10.5). More specifically, nutrient-driven algal blooms may contribute to ocean acidification possibly through increased decomposition which lowers dissolved oxygen levels in the water column and contributes to lower pH. Such nutrientenhanced acidification can also be exacerbated by warming (associated with increased microbial respiration) and changes in buffering capacity (alkalinity) of freshwater inputs (ISA, Appendix 10, section 10.5).

17

# 4.2.2.1.2 Aquatic Ecosystem Sensitivity

18 Current evidence continues to support the conclusions of the previous review regarding 19 ecosystem sensitivity to nutrient enrichment. Freshwater systems that are likely to be most 20 impacted by nutrient enrichment due to atmospheric deposition of N are remote, oligotrophic, 21 high-elevation water bodies with limited local nutrient sources and with low N retention 22 capacity. Freshwater systems sensitive to N nutrient enrichment include those in the Snowy 23 Range in Wyoming, the Sierra Nevada Mountains, and the Colorado Front Range. A portion of 24 these lakes and streams where effects are observed are in Class I wilderness areas (Williams et 25 al., 2017a; Clow et al., 2015; Nanus et al., 2012).

26 Recent research also supports the 2008 ISA findings that N limitation is common in 27 oligotrophic waters in the western U.S. (Elser et al., 2009b; Elser et al., 2009a). Shifts in nutrient 28 limitation, from N limitation, to between N and P limitation, or to P limitation, were reported in 29 some alpine lake studies reviewed in the 2008 ISA and in this review. Since the 2008 ISA, 30 several meta-analyses have reported an increase in P deposition to water bodies (Stoddard et al., 31 2016; Brahney et al., 2015; Tipping et al., 2014) and highlight the need to account for how 32 sustained P deposition can modify the effects of anthropogenically emitted N deposition on 33 productivity. Even small inputs of N in these water bodies can increase nutrient availability or 34 alter the balance of N and P, which can stimulate growth of primary producers and lead to

35 changes in species richness, community composition, and diversity.

1 The relative contribution of N deposition to total N loading varies among waterbodies.

2 For example, atmospheric deposition is generally considered to be the main source of new N

3 inputs to most headwater stream, high-elevation lake, and low-order stream watersheds that are

- 4 far from the influence of other N sources like agricultural runoff and wastewater effluent (ISA,
- 5 section ES5.2). In other fresh waterbodies, however, agricultural practices and point source
- 6 discharges have been estimated to be larger contributors (ISA, Appendix 7, section 7.1.1.1).

7 Since the 2008 ISA, several long-term monitoring studies in the Appalachian Mountains, 8 the Adirondacks, and the Rocky Mountains have reported temporal patterns of declines in 9 surface water NO<sub>3</sub><sup>-</sup> concentration corresponding to declines in atmospheric N deposition (ISA, 10 Appendix 9, section 9.1.1.2). Declines in basin wide NO<sub>3</sub><sup>-</sup> concentrations have also been reported 11 for the nontidal Potomac River watershed and attributed to declines in atmospheric N deposition 12 (ISA, Appendix 7, section 7.1.5.1). A study of water quality monitoring in a watershed in Rocky 13 Mountain National Park has also reported reductions in stream water NO3<sup>-</sup> concentrations of 14 more than 40% from peak concentrations in the mid-2000s, which corresponded to decreases in

15 NOx emissions and estimated N deposition (ISA, Appendix 7, section 7.1.5.1).

16 In estuarine and near coastal systems, the prevalence and health of submerged aquatic 17 vegetation (SAV) has been identified as a biological indicator for estuarine condition (ISA, 18 Appendix 10, section 10.2.5). Previously available evidence indicated the role of N loading in 19 SAV declines in multiple U.S. estuaries. Newly available studies have reported findings of 20 increased SAV populations in two tributaries of the Chesapeake Bay corresponding to reduction 21 in total N loading from all sources since 1990 (ISA, Appendix 10, section 10.2.5). The newly 22 available studies also identify other factors threatening SAV, including increasing temperature 23 related to climate change (ISA, Appendix 10, section 10.2.5).

24 Estimates of the relative contribution of atmospheric deposition to total N loading of 25 estuarine systems vary, with analyses based on data extending across the past two to three decades estimating that most estuaries receive 15-40% of N inputs from atmospheric sources 26 27 (ISA, section ES5.2; ISA, section 7, section 7.2.1). In coastal areas, N sources may include 28 atmospheric deposition to the water surface, coastal upwelling from oceanic waters, and 29 transport from watersheds. Freshwater inflows to estuaries often transport N from agriculture, 30 urban, wastewater, and atmospheric deposition sources (ISA, IS2.2.2; ISA, Appendix 7, section 31 7.2.1).

With regard to wetland sensitivity to N deposition, in general, those wetlands receiving a larger fraction of their total water budget in the form of precipitation are more sensitive to the effects of N deposition. The relative contribution of atmospheric deposition to total wetland N loading varies with wetland type, with bogs receiving the greatest contribution and accordingly being most vulnerable to nutrient enrichment effects of N deposition (ISA, Appendix 11, section

1 11.1). For example, bogs (70-100% of hydrological input from rainfall) are more sensitive to N 2 deposition than fens (55–83% as rainfall), which are more sensitive than coastal wetlands 3 (10-20% as rainfall) (ISA, Appendix 11, section 11.10). Nearly all N loading to ombrotrophic 4 bogs<sup>5</sup> comes from atmospheric deposition because precipitation is the only source of water to 5 these wetlands. For freshwater fens, marshes, and swamps, inputs from ground and surface water 6 are often of similar order of magnitude as that from precipitation. Similarly, estuarine and coastal 7 wetlands receive water from multiple sources that include precipitation, ground and/or surface 8 water, and marine and/or estuarine waters (ISA, Appendix 11, section 11.1).

9

# 4.2.2.1.3 Key Uncertainties

10 Models are used extensively to simulate the movement of N to sensitive receptors in 11 aquatic ecosystems, and to estimate indicators of eutrophication risk. In the case of estuarine and 12 near-coastal systems, the models are hydrodynamically complex and due to the need for inputs 13 particular to the waterbody to which they are applied, tend to be site specific (NRC, 2000; ISA, 14 Appendix 7, section 7.2.8.2). Other model uncertainties may arise from the difficulties in 15 disentangling N input sources and apportioning the source of N in the ecosystem correctly. This 16 leads to uncertainty in the role of atmospheric deposition in the N driven effects that are 17 observed.

18 Several uncertainties contribute to estimates of N deposition associated with different 19 types of water body responses. These include a difficulty in estimating dry deposition of gaseous 20 and particulate N to complex surfaces; extremely limited data, particularly for arid, mountainous 21 terrain; and difficulties estimating deposition in areas with high snowfall, cloud water or fog 22 (ISA, Appendix 9, section 9.5; Pardo et al., 2011). For example, "N deposition estimates at high-23 elevation sites such as those in the Rocky and Sierra Nevada mountains are associated with 24 considerable uncertainty, especially uncertainty for estimates of dry deposition" (ISA, Appendix 25 9, p. 9-44; Williams et al., 2017b). For estimates of N deposition associated with other sensitive 26 responses, such as shifts in phytoplankton communities in high-elevation lakes, "N deposition 27 model bias may be close to, or exceed, predicted critical load values" (ISA, Appendix 9, p. 9-44; 28 Williams et al., 2017b).

29

## 4.2.2.2 Terrestrial Ecosystems

It is long established that N enrichment of terrestrial ecosystems increases plant
 productivity (ISA, Appendix 6, section 6.1). Building on this, the currently available evidence,

32 including evidence that is longstanding, is sufficient to infer a causal relationship between N

<sup>&</sup>lt;sup>5</sup> Ombrotrophic bogs develop in areas where drainage is impeded and precipitation exceeds evapotranspiration (ISA, Appendix 11, section 11.1).

1 deposition and the alteration of the physiology and growth of terrestrial organisms and the

- 2 productivity of terrestrial ecosystems (ISA, section 5.2 and Appendix 6, section 6.2). Responsive
- 3 ecosystems include those that are N limited and/or contain species that have evolved in nutrient-
- 4 poor environments. Because N limitation is common, most terrestrial ecosystems are responsive
- 5 to increased levels of N. In these ecosystems the N-enrichment changes in plant physiology and
- 6 growth rates vary among species, with species that are adapted to low N supply being readily
- 7 outcompeted by species that have higher N demand. Because over evolutionary time, low N
- 8 conditions were much more common than high N conditions, there are many more species
- 9 adapted to low N conditions compared with species adapted to high N conditions. Thus, there is
- 10 often a net loss of species as ecosystems receive more N, whether from atmospheric deposition
- 11 or otherwise. In this manner, the relative representation of different species may be altered, and
- 12 some species may be eliminated altogether, such that community composition is changed and
- 13 species diversity declines (ISA, Appendix 6, sections 6.3.2 and 6.3.8). The currently available
- 14 evidence in this area is sufficient to infer a causal relationship between N deposition and the
- 15 alteration of species richness, community composition, and biodiversity in terrestrial ecosystems
- 16 (ISA, section IS.5.3 and Appendix 6, section 6.3).
- 17

# 4.2.2.2.1 Nature of Effects and New Evidence

18 Previously available evidence described the role of N deposition in changing soil carbon 19 and N pools and fluxes, as well as altering plant and microbial growth and physiology in an array 20 of terrestrial ecosystems. This evidence supported our understanding in the last review of how N 21 deposition influences plant physiology, growth, and terrestrial ecosystem productivity. The 22 newly available evidence confirms these conclusions and improves our understanding of the 23 mechanisms that link N deposition and biogeochemistry in terrestrial ecosystems. The new 24 evidence supports a more detailed understanding of how N influences terrestrial ecosystem 25 growth and productivity; community composition and biodiversity in sensitive ecosystems (ISA, 26 Appendix 6, section 6.2.1).

27 A supply of N is essential for plant growth and, as was clear in the last review, N 28 availability is broadly limiting for productivity in many terrestrial ecosystems (ISA, Appendix 6, 29 section 6.2.1). Accordingly, N additions contribute to increased productivity and can alter 30 biodiversity. Eutrophication, one of the mechanisms by which this can occur, comprises multiple 31 effects that include changes to the physiology of individual organisms, alteration of the relative 32 growth and abundance of various species, transformation of relationships between species, and 33 indirect effects on availability of essential resources other than N, such as light, water, and 34 nutrients (ISA, Appendix 6, section 6.2.1).

1 The currently available evidence base for the terrestrial ecosystem effects of N 2 enrichment, including eutrophication, includes studies in a wide array of systems, including 3 forests (tropical, temperate, and boreal), grasslands, arid and semi-arid scrublands, and tundra 4 (ISA, Appendix 6). The organisms affected include trees, herbs and shrubs, and lichen, as well as 5 fungal, microbial, and arthropod communities. As recognized in section 4.1 above, lichen 6 communities, which have important roles in hydrologic cycling, nutrient cycling, and as sources 7 of food and habitat for other species, are also affected by atmospheric N (ISA, Appendix 6). The 8 recently available studies on the biological effects of added N in terrestrial ecosystems include 9 investigations of plant and microbial physiology, long-term ecosystem-scale N addition 10 experiments, regional and continental-scale monitoring studies, and syntheses.

11 The previously available evidence included N addition studies in the U.S. and N 12 deposition gradient studies in Europe showing reduced species richness and altered community 13 composition for grassland plants, forest understory plants, and mycorrhizal fungi (soil fungi that 14 have a symbiotic relationship with plant roots) (ISA, Appendix 6, section 6.3). Since 2008, new 15 research techniques have been developed to understand community composition, additional 16 communities have been surveyed, and new studies have made it possible to isolate the influence 17 of N deposition from other environmental factors. In addition, new evidence has been developed 18 for forest communities indicating that N deposition alters the physiology and growth of overstory 19 trees, and that N deposition has the potential to change the community composition of forests 20 (ISA, Appendix 6, section 6.6). Recent studies on forest trees include analyses of long-term 21 forest inventory data collected from across the U.S. and Europe (ISA, Appendix 6, section 22 6.2.3.1). New research also expands the understanding that N deposition can alter the 23 physiology, growth, and community composition of understory plants, lichens, mycorrhizal 24 fungi, soil microorganisms, and arthropods (ISA, Appendix 6, section 6.2.3 and 6.3.3). 25 The recent evidence includes findings of variation in forest understory and non-forest 26 plant communities with atmospheric N deposition gradients in the U.S. and in Europe. For 27 example, gradient studies in Europe have found higher N deposition to be associated with forest 28 understory plant communities with more nutrient-demanding and shade-tolerant plant species 29 (ISA, Appendix 6, section 6.3.3.2). A recent gradient study in the U.S. found forest understory 30 species richness to be highly dependent on soil pH, with species richness declining at N 31 deposition rates >11.6 kg N/ha/yr at sites with low soil pH but not having a negative effect, up to 32 deposition levels of 20 kg N/ha/yr, at the sites with basic soils (ISA, Appendix 6, section 33 6.3.3.2).

Among the new studies are investigations of effects of N on mycorrhizal fungi and
 lichens. Studies indicate that increased N in forest systems can result in changes in mycorrhizal
 community composition (ISA, Appendix 6, section 6.2). Forest microbial biomass and

1 community composition can also be affected, which can contribute to impacts on arthropod 2 communities (ISA, Appendix 6, section 6.3.3.4). Recent evidence includes associations of 3 variation in lichen community composition with N deposition gradients in the U.S. and Europe, 4 (ISA, Appendix 6, section 6.2.6; Table 6-23). Differences in lichen community composition have 5 been attributed to atmospheric N pollution in forests throughout the West Coast, in the Rocky 6 Mountains, and in southeastern Alaska. Differences in epiphytic lichen growth or physiology 7 have been observed along atmospheric N deposition gradients in the highly impacted area of 8 southern California, and also in more remote locations such as Wyoming and southeastern 9 Alaska (ISA, Appendix 6, section 6.3.7). Historical deposition may play a role in observational 10 studies of N deposition effects, complicating the disentangling of responses that may be related 11 to more recent N loading.

12 Newly available findings from N addition experiments expand on the understanding of 13 mechanisms linking changes in plant and microbial community composition to increased N 14 availability. Such experiments in arid and semi-arid environments indicate that competition for 15 resources such as water may exacerbate the effects of N addition on diversity (ISA, Appendix 6, 16 section 6.2.6). A 25-year experiment with N additions ranging from 10 to 95 kg N/ha-yr (and 17 background wet deposition of N estimated at 6 kg N/ha-yr) observed grassland composition to 18 change from a high-diversity, native-dominated state to a low-diversity, non-native dominated 19 state (ISA, Appendix 6, section 6.3.5). The newly available evidence also includes studies in arid 20 and semiarid ecosystems, particularly in southern California, that have reported changes in plant 21 community composition, in the context of a long history of significant N deposition, with fewer 22 observations of plant species loss or changes in plant diversity (ISA, Appendix 6, section 6.3.6).

23

## 4.2.2.2.2 Terrestrial Ecosystem Sensitivity

24 In general, most terrestrial ecosystems are N limited and, consequently, sensitive to 25 effects related to N enrichment (ISA, Appendix 6, section 6.3.8). Factors identified as governing 26 the sensitivity of terrestrial ecosystems to nutrient enrichment from N deposition include "the 27 rates of N deposition, degree of N limitation, ecosystem productivity, elevation, species 28 composition, length of growing season, and soil N retention capacity" (ISA, Appendix 6, p. 6-29 162). One example is that of alpine tundra ecosystems, which: (1) are typically strongly N 30 limited, contain vegetation adapted to low N availability; (2) often have thin soils with limited N 31 retention capacity; and (3) have short growing seasons (ISA, Appendix 6, section 6.3.8). Given 32 the evidence regarding sensitivity of lichens and ectomycorrhizal fungi to N enrichment effects, 33 it may be that ecosystems containing a large number and/or diversity of these organisms, such as 34 temperate and boreal forests and alpine tundra, could be considered particularly sensitive to N 35 deposition (ISA, Appendix 6, sections 6.2.3.2, 6.2.3.3, 6.2.4, and 6.3.8).

1 In the currently available evidence, studies conducted in grassland and coastal sage shrub 2 communities, and in arid ecosystems, such as the Mojave Desert, indicate sensitivity of those 3 communities. For example, N addition studies in Joshua Tree National Park have reported losses 4 in forb species richness (which make up most of the grassland biodiversity), greater growth of 5 grass species (which make up the majority of grassland biomass), and changes in reproductive 6 rates. Accordingly, the N limitation in grasslands and the dominance by fast-growing species that 7 can shift in abundance rapidly (in contrast to forest trees) contribute to an increased sensitivity of 8 grassland ecosystems to N inputs (ISA, Appendix 6, section 6.3.6). Studies in southern 9 California coastal sage scrub communities, including studies of the long-term history of N 10 deposition, which was appreciably greater in the past than recent rates, indicate impacts on 11 community composition and species richness in these ecosystems (ISA, Appendix 6, sections 12 6.2.6 and 6.3.6). In summary, the ability of atmospheric N deposition to override the natural 13 spatial heterogeneity in N availability in arid ecosystems, such as the Mojave Desert and CSS 14 ecosystems in southern California, makes these ecosystems sensitive to N deposition (ISA, 15 Appendix 6, section 6.3.8).

16 The current evidence includes relatively few studies of N enrichment recovery in 17 terrestrial ecosystems. Among N addition studies assessing responses after cessation of 18 additions, it has been observed that soil nitrate and ammonium concentrations recovered to levels 19 observed in untreated controls within 1 to 3 years of the cessation of additions, but soil processes 20 such as N mineralization and litter decomposition were slower to recover (ISA, Appendix 6, 21 section 6.3.2; Stevens, 2016). A range of recovery times have been reported for mycorrhizal 22 community composition and abundance from a few years in some systems to as long as 28 or 48 23 years in others (ISA, Appendix 6, section 6.3.2; Stevens, 2016; Emmett et al., 1998; Strengbom 24 et al., 2001). An N addition study in the midwestern U.S. observed that plant physiological 25 processes recovered in less than 2 years, although grassland communities were slower to recover 26 and still differed from controls 20 years after the cessation of N additions (ISA, Appendix 6, 27 section 6.3.2; Isbell et al., 2013b).

28

## 4.2.2.2.3 Key Uncertainties

Just as there are uncertainties associated with estimating N deposition associated with ecological responses in aquatic systems (as summarized in section 4.2.2.1.3 above), such uncertainties exist with terrestrial ecosystem analyses. For example, regarding wet deposition measurements, there are uncertainties associated with monitoring instrumentation and measurement protocols, as well as limitations in the spatial extent of existing monitoring networks, especially in remote areas. Given limitations in our ability to estimate dry deposition, estimates are often based on model predictions, for which there are various sources of

1 uncertainty, including model formulation and inputs for the simulation of chemistry and

- 2 transport processes. Other uncertainties are associated with an incomplete understanding of the
- 3 underlying scientific processes influencing atmospheric deposition that are not possible to
- 4 quantify. For example, uncertainties associated with deposition estimates (that may be utilized in
- 5 observational studies) include those associated with simulating effects of the tree canopy on NO<sub>x</sub>
- 6 (including both bidirectional gas exchange and canopy reactions), bidirectional exchange of NH<sub>3</sub>
- 7 with biota and soils, and processes determining transference ratios that relate average
- 8 concentration to deposition. (ISA, section IS.14.1.3).
- 9 There is also uncertainty with regard to the relative importance of different N species in 10 effects of N enrichment on terrestrial ecosystem [ISA, Appendix 6, section 6.3.2]. Although
- 11 there are few direct analyses comparing the impacts of oxidized and reduced forms of N
- 12 deposition on biodiversity, it is plausible that  $NO_3^-$  may be less likely to accumulate in soil, with
- 13 associated effects, due to its greater tendency to be more readily lost to both leaching and
- 14 denitrification than NH<sub>4</sub><sup>+</sup> (ISA, Appendix 6, section 6.3.2). Further, while multiple meta-
- 15 analyses have generally not reported differences in the relationship of different N forms with
- 16 ecological and biogeochemical endpoints, such as plant productivity or microbial biomass,
- 17 several individual studies have observed differential effects on diversity of NH<sub>4</sub><sup>+</sup> versus NO<sub>3</sub><sup>-</sup>
- 18 additions. For example, an experiment involving a nutrient-poor, Mediterranean site found that
- 19 while an NH<sub>4</sub><sup>+</sup> addition (40 kg N/ha/yr) increased plant richness, addition of the same amount of
- 20 N comprised of half  $NH_4^+$  and half  $NO_3^-$  did not (ISA, Appendix 6, section 6.3.2).
- With regard to ecological responses and impacts of concern, there are several key areas of uncertainty. In observational studies, in addition to uncertainty regarding the role of historical deposition, other confounding factors such as drought and ozone may also contribute to impacts of concern. Further, there is wide variability in the response of plants to nitrogen inputs and the impacts of spatially variable factors such as climate, geology and past deposition on that response is generally unknown. Spatially, variation in biological and biogeochemical processes imposed by climate, geology, biota, and other environmental factors may affect observed associations of ecological metrics with deposition metrics.
- associations of ecological metrics with deposition metrics.
  Uncertainties also relate to time scales and lags. For example, while atmospheric
  deposition responds dynamically to shifts in emissions and weather patterns, ecological
  processes react to environmental stress at a variety of timescales, which due to intervening
  ecosystem processes usually lag changes in deposition. There are also uncertainties related to the
  role of historic patterns of deposition in ecosystem effects initially attributed to recent gradients
  in deposition. These may loom larger for geographic regions, such as the northeastern U.S. or
- 35 southern California that have long and geographically extensive histories of elevated N

4-26

36 deposition.

#### 1 4.2.3 Other Effects

Additional categories of effects for which the current evidence is sufficient to infer causal
relationships include changes in mercury methylation processes in freshwater ecosystems,
changes in aquatic biota due to sulfide phytotoxicity, and ecological effects from PM deposition
(ISA, Table IS-1).

6

#### 4.2.3.1 Mercury Methylation

7 The current evidence, including that newly available in this review, is sufficient to infer a 8 causal relationship between S deposition and the alteration of Hg methylation in surface water, 9 sediment, and soils in wetland and freshwater ecosystems. The process of mercury methylation is 10 influenced in part by surface water  $SO_4^{2-}$  concentrations, as well as the presence of mercury. 11 Accordingly, in waterbodies where mercury is present, S deposition, particularly that associated 12 with SOx has a role in production of methylmercury, which contributes to methylmercury 13 accumulation in fish (ISA, Appendix 12, section 12.8).

14 Newly available evidence has improved our scientific understanding of the types of 15 organisms involved in the methylation process, as well as the environments in which they are 16 found. Studies have also identified additional areas within the U.S. containing habitats with 17 conditions suitable for methylation, and species that accumulate methylmercury (ISA, Appendix 18 12, section 12.3). The evidence also contributes to our understanding of factors that can 19 influence the relationship between atmospheric S deposition and methyl mercury in aquatic 20 systems; such factors include oxygen content, temperature, pH, and carbon supply, which 21 themselves vary temporally, seasonally, and geographically (ISA, Appendix 12, section 12.3).

22

## 4.2.3.2 Sulfide Toxicity

23 The evidence newly available in this review regarding non-acidifying sulfur effects on 24 biota expands upon that available for the 2008 ISA. The currently available evidence is sufficient 25 to infer a new causal relationship between S deposition and changes in biota due to sulfide 26 phytotoxicity, including alteration of growth and productivity, species physiology, species 27 richness, community composition, and biodiversity in wetland and freshwater ecosystems (ISA, 28 section IS.9). The currently available evidence indicates that the presence of sulfide interferes 29 with nutrient uptake in roots of plants in wetlands and other fresh waterbodies. Studies also 30 report that elevated sulfide can result in decreased seed mass, seed viability, seedling emergence 31 rates, decreased seedling height, decreased seedling survival rates, and reductions in total plant 32 cover, all which have the potential to contribute to shifts in plant community composition (ISA, 33 Appendix 12, section 12.2.3). Sulfur deposition can contribute to sulfide and associated 34 phytotoxicity in freshwater wetlands and lakes. Recently available studies indicate that sulfide 35 toxicity can occur in wetland habitats and suggests that sulfide toxicity can determine plant

1 community composition in freshwater wetlands. These studies indicate sulfide toxicity to have 2 occurred in multiple wetland ecosystems in North America (ISA, Appendix 12, sections12.2.3 3 and 12.7.3).

4

#### 4.2.3.3 Ecological Effects of PM Other Than N and S Deposition

5 Particulate matter includes a heterogeneous mixture of particles differing in origin, size, 6 and chemical composition. In addition to N and S and their transformation products, other PM 7 components, such as trace metals and organic compounds are also deposited to ecosystems and 8 may affect biota. Material deposited onto leaf surfaces can alter leaf processes and PM 9 components deposited to soils and waterbodies may be taken up into biota, with the potential for 10 effects on biological and ecosystem processes. The currently available evidence is sufficient to 11 infer a likely causal relationship between deposition of PM and a variety of effects on individual 12 organisms and ecosystems (ISA, Appendix 15, section 15.1).

13 The effects of PM on ecological receptors can be both chemical and physical, and 14 particles that elicit effects on ecological receptors vary by size, origin, and chemical 15 composition. Although in some limited cases, effects have been attributed to particle size (e.g., 16 soiling of leaves by large coarse particles near industrial facilities or unpaved roads), ecological 17 effects of PM have been largely attributed more to particle composition (Grantz et al., 2003; ISA, 18 Appendix 15, section 15.2). For example, exposure to a given mass-per-volume or -mass 19 concentration may result in quite different ecological effects depending on the PM components. 20 Depending on concentration, trace metals, some of which are biologically essential, can be toxic 21 in large amounts (ISA, Appendix 15, section 15.3.1). Depending on conditions, deposited PM 22 has been associated with effects on vegetation including effects on plant surfaces, foliar uptake 23 processes, gas exchange, physiology, growth, and reproduction. The evidence largely comes

24 from studies involving elevated concentrations such as near industrial areas or historically

25 polluted cities (ISA, Appendix 15, section 15.4). Recent assays have supported previously

26 available evidence that toxicity relates more to chemical components than total mass.

27 Additionally recent experiments have suggested that PM deposition can influence responses in

28 microbial communities (ISA, Appendix 15, section 15.8). Quantifying relationships between

29 ambient air concentrations of PM and ecosystem response are difficult and uncertain.

# 30

4.3 PUBLIC WELFARE IMPLICATIONS

31 The public welfare implications of the evidence regarding S and N related welfare effects 32 are dependent on the type and severity of the effects, as well as the extent of the effect at a 33 particular biological or ecological level of organization or spatial scale. We discuss such factors 34 here in light of judgments and conclusions made in NAAQS reviews regarding effects on the 35 public welfare.

1 As provided in section 109(b)(2) of the CAA, the secondary standard is to "specify a 2 level of air quality the attainment and maintenance of which in the judgment of the 3 Administrator ... is requisite to protect the public welfare from any known or anticipated adverse 4 effects associated with the presence of such air pollutant in the ambient air." The secondary 5 standard is not meant to protect against all known or anticipated welfare effects related to oxides 6 of N and S, and particulate matter, but rather those that are judged to be adverse to the public 7 welfare, and a bright-line determination of adversity is not required in judging what is requisite 8 (78 FR 3212, January 15, 2013; 80 FR 65376, October 26, 2015; see also 73 FR 16496, March 9 27, 2008). Thus, the level of protection from known or anticipated adverse effects to public 10 welfare that is requisite for the secondary standard is a public welfare policy judgment made by 11 the Administrator. The Administrator's judgment regarding the available information and 12 adequacy of protection provided by an existing standard is generally informed by considerations 13 in prior reviews and associated conclusions.

14 15

# Is there newly available information relevant to consideration of the public welfare implications of S and N deposition-related welfare effects?

16 There is a large body of newly available evidence regarding the impacts of S and N 17 deposition on biological/ecological resources across a wide range of effects that can be used to 18 help inform public welfare considerations. The categories of effects identified in the CAA to be 19 included among welfare effects are in fact quite diverse,<sup>6</sup> and among these categories, any single 20 category includes many different types of effects that are of broadly varying specificity and level 21 of resolution. For example, effects on vegetation and effects on animals are categories identified 22 in CAA section 302(h), and the ISA recognizes numerous effects of N and S deposition at the 23 organism, population, community, and ecosystem level, as summarized in sections 4.1 and 4.2 24 above (ISA, sections IS.5 to IS.9). The significance of each type of effect with regard to potential 25 effects on the public welfare depends on the type and severity of effects, as well as the extent of 26 such effects on the affected environmental entity, and on the societal use of the affected entity 27 and the entity's significance to the public welfare. Such factors have been considered in the 28 context of judgments and conclusions made in some prior reviews regarding public welfare 29 effects. As noted in the last review of the secondary NAAQS for NOx and SOx, while the CAA 30 section 302(h) lists a number of welfare effects, "these effects do not define public welfare in

31 and of themselves" (77 FR 20232, April 3, 2012).

<sup>&</sup>lt;sup>6</sup> Section 302(h) of the CAA states that language referring to "effects on welfare" in the CAA "includes, but is not limited to, effects on soils, water, crops, vegetation, manmade materials, animals, wildlife, weather, visibility, and climate, damage to and deterioration of property, and hazards to transportation, as well as effects on economic values and on personal comfort and well-being" (CAA section 302(h)).

1 In the context of secondary NAAOS decisions for ozone, judgments regarding public 2 welfare significance have given particular attention to effects in areas with special federal protections (such as Class I areas),<sup>7</sup> and lands set aside by states, tribes and public interest groups 3 to provide similar benefits to the public welfare (73 FR 16496, March 27, 2008; 80 FR 65292, 4 5 October 26, 2015).<sup>8</sup> For example, in the 2015 O<sub>3</sub> NAAQS review, the EPA recognized the "clear 6 public interest in and value of maintaining these areas in a condition that does not impair their 7 intended use and the fact that many of these lands contain O<sub>3</sub>-sensitive species" (73 FR 16496, 8 March 27, 2008). Judgments regarding effects on the public welfare can depend on the intended 9 use for, or service (and value) of, the affected vegetation, ecological receptors, ecosystems and 10 resources and the significance of that use to the public welfare (73 FR 16496, March 27, 2008: 11 80 FR 65377, October 26, 2015). Uses or services provided by areas that have been afforded 12 special protection can flow in part or entirely from the vegetation that grows there or other 13 natural resources. Ecosystem services range from those directly related to the natural functioning 14 of the ecosystem to ecosystem uses for human recreation or profit, such as through the 15 production of lumber or fuel (Costanza et al., 2017; ISA, section IS.5.1). The spatial, temporal, 16 and social dimensions of public welfare impacts are also influenced by the type of service 17 affected. For example, a national park can provide direct recreational services to the thousands of 18 visitors that come each year, but also provide an indirect value to the millions who may not visit 19 but receive satisfaction from knowing it exists and is preserved for the future (80 FR 65377, 20 October 26, 2015). 21 In the last review of the secondary NAAQS for NOx and SOx, ecosystem services were 22 discussed as a method of assessing the magnitude and significance to the public of resources

23 affected by ambient air concentrations of oxides of nitrogen and sulfur and deposition in

sensitive ecosystems (77 FR 20232, April 3, 2012). That review recognized that although there is

25 no specific definition of adversity to public welfare, one paradigm might involve ascribing public

<sup>&</sup>lt;sup>7</sup> Areas designated as Class I include all international parks, national wilderness areas which exceed 5,000 acres in size, national memorial parks which exceed 5,000 acres in size, and national parks which exceed 6,000 acres in size, provided the park or wilderness area was in existence on August 7, 1977. Other areas may also be Class I if designated as Class I consistent with the CAA.

<sup>&</sup>lt;sup>8</sup> For example, the fundamental purpose of parks in the National Park System "is to conserve the scenery, natural and historic objects, and wild life in the System units and to provide for the enjoyment of the scenery, natural and historic objects, and wild life in such manner and by such means as will leave them unimpaired for the enjoyment of future generations" (54 U.S.C. 100101). Additionally, the Wilderness Act of 1964 defines designated "wilderness areas" in part as areas "protected and managed so as to preserve [their] natural conditions" and requires that these areas "shall be administered for the use and enjoyment of the American people in such manner as will leave them unimpaired for future use and enjoyment as wilderness, and so as to provide for the protection of these areas, [and] the preservation of their wilderness character …" (16 U.S.C. 1131 (a) and (c)). Other lands that benefit the public welfare include national forests which are managed for multiple uses including sustained yield management in accordance with land management plans (see 16 U.S.C. 1600(1)-(3); 16 U.S.C. 1601(d)(1)).

1 welfare significance to disruptions in ecosystem structure and function. The concept of

2 considering the extent to which a pollutant effect will contribute to such disruptions has been

3 used broadly by the EPA in considering effects. An evaluation of adversity to public welfare

4 might also consider the likelihood, type, magnitude, and spatial scale of the effect, as well as the

potential for recovery and any uncertainties relating to these considerations (77 FR 20218, April
3, 2012).

7 The types of effects on aquatic and terrestrial ecosystems discussed in sections 4.2 and 8 4.3 above differ with regard to aspects important to judging their public welfare significance. For 9 example, in the case of effects on timber harvest, such judgments may consider aspects such as 10 the heavy management of silviculture in the U.S., while judgments for other categories of effects 11 may generally relate to considerations regarding natural areas, including specifically those areas 12 that are not managed for harvest. For example, effects on tree growth and survival have the 13 potential to be significant to the public welfare through impacts in Class I and other areas given 14 special protection in their natural/existing state, although they differ in how they might be 15 significant.

16 In this context, it may be important to consider that S and N deposition-related effects, 17 such as changes in growth and survival of plant and animal species, could, depending on severity, extent, and other factors, lead to effects on a larger scale including changes in overall 18 19 productivity and altered community composition (ISA, section IS.2.2.1 and Appendices 5, 6, 8, 20 9, and 10). Further, effects on individual species could contribute to impacts on community 21 composition through effects on growth and reproductive success of sensitive species in the 22 community, with varying impacts to the system through many factors including changes to 23 competitive interactions (ISA, section IS.5.2 and Appendix 6, section 6.3.2). Impacts on some of 24 these characteristics (e.g., forest or forest community composition) may be considered of greater 25 public welfare significance when occurring in Class I or other protected areas, due to the value 26 that the public places on such areas. Other ecosystem services that can be affected are summarized below in Figure 4-3<sup>9</sup> (ISA, Appendix 14). In considering such services in past 27 28 reviews for secondary standards for other pollutants (e.g., O<sub>3</sub>), the Agency has given particular 29 attention to effects in natural ecosystems, indicating that a protective standard, based on 30 consideration of effects in natural ecosystems in areas afforded special protection, would also 31 "provide a level of protection for other vegetation that is used by the public and potentially 32 affected by O<sub>3</sub> including timber, produce grown for consumption and horticultural plants used

33 for landscaping" (80 FR 65403, October 26, 2015).

<sup>&</sup>lt;sup>9</sup> The articulation of welfare effects in Figure 4-3 is intended to reflect the ISA causal determinations in an easier to comprehend manner that also illustrates connections among effects.

1 However, available information does not yet provide a framework that can specifically tie 2 changes in a biological or ecological indicator (e.g., lichen abundance) from deposition and 3 broad effects on the public welfare. This gap creates uncertainties when considering the public 4 welfare implications of some biological or geochemical responses to ecosystem acidification or 5 N enrichment, and accordingly judgments on the potential for public welfare significance. That 6 notwithstanding, while shifts in species abundance or composition of various ecological 7 communities may not be easily judged with regard to public welfare significance, at some level, 8 such changes, especially if occurring broadly in specially protected areas, where the public can 9 be expected to place high value, might reasonably be concluded to impact the public welfare. An 10 additional complexity in the current review is the current air quality and associated deposition 11 within the context of a longer history that included appreciably greater deposition in the middle 12 of the last century, the environmental impacts of which may remain. 13 In summary, several considerations are recognized as important to judgments on the 14 public welfare significance of the array of welfare effects at different exposure conditions. These 15 include uncertainties and limitations that must be taken into account regarding the magnitude of 16 key effects that might be concluded to be adverse to ecosystem health and associated services. 17 Additionally, there are numerous locations vulnerable to public welfare impacts from S or N 18 deposition-related effects on terrestrial and aquatic ecosystems and their associated services. 19 Other important considerations include the exposure circumstances that may elicit effects and the 20 potential for the significance of the effects to vary in specific situations due to differences in 21 sensitivity of the exposed species, the severity and associated significance of the observed or 22 predicted effect, the role that the species plays in the ecosystem, the intended use of the affected

23 species and its associated ecosystem and services, the presence of other co-occurring

24 predisposing or mitigating factors, and associated uncertainties and limitations.



Figure 4-3. Potential effects on the public welfare of ecological effects of N Oxides, SOx and PM.

2 3

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5 EXPOSURE CONDITIONS ASSOCIATED WITH EFFECTS

3 In this review, we consider two categories of exposure conditions associated with welfare effects. The first is the less complex consideration of the direct exposures to pollutants in 4 5 ambient air, which were the focus in the establishment of the standards. The second is the more 6 complex consideration exposures related to atmospheric deposition associated with the pollutants 7 in ambient air. In our consideration in this chapter of exposure conditions associated with effects, we have generally addressed the two categories in separate sections. 8 Section 5.1 discusses the currently available information related to consideration of 9 10 exposure concentrations associated with direct welfare effects of nitrogen and sulfur oxides and PM in ambient air. This is done in the context of the following overarching question: 11 To what extent does the available evidence include quantitative exposure and 12 response information that can inform judgments on air exposures of concern and 13 accordingly, the likelihood of occurrence of such effects in response to air quality 14 that meets the current standards? 15 Sections 5.2 through 5.4 address the more complex consideration of deposition-related 16 17 exposures, which was a major focus in the 2012 review of the secondary standards for oxides of sulfur and nitrogen. In this regard, we consider the following policy-relevant question: 18 To what extent does the available evidence provide quantitative linkages of S oxides, 19 N oxides and/or PM deposition and effects that can inform judgments on deposition 20 levels of concern and accordingly, the likelihood of occurrence of such effects in 21 response to air quality that meets the current standards? 22 There is wide variation in the extent and level of detail of the evidence available to 23 describe the ecosystem characteristics (e.g., physical, chemical, and geological characteristics, as 24 25 well as atmospheric deposition history) that influence the degree to which deposition of N and S 26 associated with the oxides of S and N and PM in ambient air elicit ecological effects. One reason 27 for this relates to the contribution of many decades of uncontrolled atmospheric deposition before the establishment of NAAQS for PM, oxides of S and oxides of N, followed by the 28 29 subsequent decades of continued deposition as standards were implemented and updated. The 30 impacts of this deposition history remain in soils of many parts of the U.S. today (e.g., in the Northeast and portions of the Appalachian Mountains in both hardwood and coniferous forests, 31 32 as well as areas in and near the Los Angeles Basin), with recent signs of recovery in some areas

33 (ISA, Appendix 4, section 4.6.1; 2008 ISA, section 3.2.1.1). This backdrop and associated site-

34 specific characteristics are among the challenges we consider in our task of identifying

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deposition targets to provide protection going forward against the array of effects for which we
 have evidence of occurrence in sensitive ecosystems as a result of the deposition of the past.
 With regard to aquatic systems, prior to the peak of S deposition levels that occurred in
 the 1970s and early 80s, surface water sulfate concentrations increased in response to S

5 deposition. Subsequently, and especially more recently, concentrations have generally decreased,

6 particularly in the Northeast. Some waterbodies, however, continue to exhibit little reduction in

acidic ions, such as in the Blue Ridge Mountains region in Virginia, where surface water SO4<sup>2-</sup>
has remained relatively stable even as emissions declined. This is an example of the competing

9 role of changes in S adsorption on soils and the release of historically deposited S from soils into

10 surface water, which some modeling has suggested will delay chemical recovery in those water

11 bodies (ISA, Appendix 7, section 7.1.2.2).

In this chapter we first consider the categories of effects for which quantitative 12 assessment approaches for atmospheric deposition are the most established and robust. In the 13 14 2012 review of the oxides of N and S, quantitative analyses relating deposition in recent times (e.g., since 2000) to ecosystem acidification, and particularly aquatic acidification, were 15 generally considered to be less uncertain and the ability of those analyses to inform NAAQS 16 policy judgments more robust than analyses related to deposition and ecosystem nutrient 17 enrichment, or eutrophication (2011 PA). While the evidence base regarding atmospheric 18 deposition and nutrient enrichment has expanded since the 2012 review, this generally remains 19 20 the case in the current review. Accordingly, the chapter addresses the quantitative information 21 available for both acidification and nutrient enrichment, but there is more quantitative 22 information and associated discussion related to ecosystem acidification, and particularly aquatic acidification. 23 24 Critical loads are frequently used in studies investigating associations between an array of

Critical loads are frequently used in studies investigating associations between an array of chemical, biological, ecological and ecosystem characteristics and a variety of N or S depositionrelated metrics.<sup>1</sup> These studies vary widely with regard to the specific ecosystem characteristics being evaluated, as well as the benchmarks selected for judging them, such as the depositionrelated metrics, their scope, method of estimation and time period. The specific details of these various factors influence the strengths and limitations for different uses and have associated

<sup>&</sup>lt;sup>1</sup> The term, critical load, which in general terms refers to an amount (or a rate of addition) of a pollutant to an ecosystem that is estimated to be at, or just below, that which would have an effect of interest, has multiple interpretations or applications (ISA, p. IS-14). This multiplicity or variety in meanings stems, at least in part, from differing judgments and associated identifications regarding the effect of interest, and judgment of its harm. There is additionally the complication of the dynamic nature of ecosystem pollutant processing and the broad array of factors that influence it. As a result, time is an important dimension, sometimes unstated, as in empirical or observational analyses, sometimes explicit, as in steady-state or dynamic modeling analyses (ISA, section IS.2.2.4).

1 uncertainties. Given the role of the PA both in focusing on the most policy relevant aspects of the

- 2 currently available information (reviewed in the 2020 and 2008 ISAs and past AQCDs) and in
- 3 clearly describing key aspects, including limitations and associated uncertainties, this document

4 is intended to reach beyond individual critical loads developed over a variety of studies and

5 ecosystems and consider the underlying study findings with regard to key aspects of the

6 environmental conditions and ecological characteristics studied.

A more quantitative variation of this approach is the methodology developed for the
analyses of aquatic systems and acidification, summarized in section 5.2.2 below. In these
analyses, the concept of a critical load is employed with steady-state modeling that relates
deposition to waterbody acid neutralizing capacity. This specific use of critical loads is explicitly
described in section 5.2.2.

While recognizing the inherent connections between watersheds and waterbodies (lakes 12 and streams), the organization of this chapter recognizes the more established state of the 13 14 information, tools and data for aquatic ecosystems for characterizing relationships between 15 atmospheric deposition and acidification and/or nutrient enrichment effects under air quality 16 associated with the current standards. Further, we recognize the relatively greater role of atmospheric deposition in aquatic acidification compared to aquatic eutrophication, to which 17 18 surface water discharges in populated watersheds have long contributed. We also note that 19 recovery of aquatic ecosystem biota from aquatic acidification may in many locations be more 20 rapid than recovery of tree populations from terrestrial acidification (Driscoll et al., 2001). 21 Therefore, with regard to deposition-related effects, we focus first on the quantitative 22 information for aquatic ecosystem effects in sections 5.2 and 5.3. Section 5.4 then discusses the 23 available evidence regarding relationships between deposition-related exposures and the 24 occurrence and severity of effects on trees and understory communities in terrestrial ecosystems.

## 25 5.1 DIRECT EFFECTS OF OXIDES OF N AND S AND OF PM IN 26 AMBIENT AIR

## 27 5.1.1 Sulfur Oxides

28 As summarized in section 4.1 above, the direct welfare effects of SOx in ambient air 29 include effects on vegetation, such as foliar injury, depressed photosynthesis and reduced growth 30 or yield. Within the recently available information are observational studies reporting increased growth in association with reductions in SO<sub>2</sub> emissions. These studies, however, do not generally 31 32 report the SO<sub>2</sub> concentrations in ambient air or account for the influence of changes in 33 concentrations of co-occurring pollutants such as ozone (ISA, Appendix 3, section 3.2). The available data that includes exposure concentrations is drawn from experimental studies or 34 35 observational studies in areas near sources, with the most studied effect being visible foliar

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1 injury to various trees and crops (ISA, Appendix 3, section 3.2; 1982 AQCD, section 8.3). Based

- 2 on controlled laboratory exposures in some early studies (assessed in the 1982 AQCD),
- 3 concentrations greater than approximately  $0.3 \text{ ppm SO}_2$  for a few hours were required to induce
- 4 slight injury in seedlings of several pine species, with sensitive species exposed in conducive
- 5 conditions being more likely to show visible injury (1982 AQCD, section 8.3). With regard to
- 6 foliar injury, the current ISA states there to be "no clear evidence of acute foliar injury below the
- 7 level of the current standard" (ISA, p. IS-37). For effects on plant productivity and growth,
- 8 studies described in the 1982 AQCD that involve experimental exposures in the laboratory have
- 9 reported depressed photosynthesis by 20% or more from one week of continuous exposure to 0.5
- 10 ppm  $SO_2$  or 3 weeks to 3 hours/day at 0.5 ppm. Few studies report yield effects from acute
- 11 exposures, with the available ones reporting relatively high concentrations. For example, a study
- 12 with soybeans reported statistically significant yield reductions (more than 10%) after a 4.2-hour
- exposure to concentrations greater than 1 ppm  $SO_2$  (1982 AQCD, section 8.3).

14 The evidence presented in the ISA also includes effects on lichen species, such as those reported in laboratory fumigation experiments that have assessed effects on photosynthesis and 15 other functions in a few lichen groups (ISA, Appendix 3, section 3.2). For example, a study of 16 two lichens in Spain by Sanz et al. (1992) found photosynthesis to be significantly depressed in 17 18 the more sensitive species after 4 to 6 hours at 0.1 ppm SO<sub>2</sub>, with recovery occurring within 2 19 hours following exposure. After two weeks, however, recovery had not occurred after significant 20 reduction in photosynthesis from six hours at 0.25 ppm. After shorter exposures to 0.25, 0.5 and 0.9 ppm, photosynthesis recovered within two weeks. After exposures to 0.9 and 1.5 ppm SO<sub>2</sub> 21 22 ppm for one to six hours, photosynthesis was significantly reduced and did not recover. The 23 second species tested was appreciably less sensitive, with photosynthesis not being affected for 24 lower exposures than six hours at a concentration of 0.5 ppm SO<sub>2</sub> (Sanz et al., 1992).

### 25 5.1.2 Nitrogen Oxides

The direct welfare effects of N oxides in ambient air include effects on plants and lichens. 26 For plants, studies reported in the ISA did not report effects on photosynthesis and growth 27 resulting from exposures of NO<sub>2</sub> concentrations below 0.1 ppm (ISA, Appendix 3, section 3.3). 28 29 For example, five days of 7-hour/day exposures of soybean plants reduced photosynthesis at 0.5 ppm, and increased photosynthesis at 0.2 ppm NO<sub>2</sub> (ISA, Appendix 3, section 3.3). Exposures to 30 0.1 ppm NO<sub>2</sub> continuously for eight weeks and for six hours/day over 28 days elicited reduced 31 growth of Kentucky blue grass and seedlings of three tree species, respectively (ISA, Appendix 32 3, section 3.3). A study of five California native grasses and forbs exposed to 0.03 ppm NO<sub>2</sub> 33 continuously for 16 weeks found no significant effects on shoot or root biomass, photosynthesis 34 35 or stomatal conductance (ISA, Appendix 3, section 3.3). Visible foliar injury has not been

1 reported to occur with NO<sub>2</sub> exposure concentrations below 0.2 ppm except for exposures of

- 2 durations lasting 100 hours or longer (ISA, Appendix 3, section 3.3). The ISA notes that for most
- 3 plants, "injury occurred in less than 1 day only when concentrations exceeded 1 ppm" (ISA,
- 4 Appendix 3, p. 3-10). The information is more limited with regard to exposures to other oxides
- 5 of N. A study involving three 4-hr exposures to 30 ppb PAN on alternating days in a laboratory
- 6 setting reported statistically significant reduction in growth of kidney bean and petunia plants
- 7 (ISA, Appendix 3, section 3.3).
- The evidence for HNO<sub>3</sub>, includes controlled exposure studies describing foliar effects on 8 9 several tree species. For example, 12-hour exposures to 50 ppb HNO<sub>3</sub> ( $\sim$ 75 µg/m<sup>3</sup>) in light, and to 200 ppb ( $\sim$ 530 µg/m<sup>3</sup>) in darkness, affected ponderosa pine needle cuticle (ISA, Appendix 3, 10 11 section 3.4). Nitric acid has also been found to deposit on and bind to the leaf or needle surfaces. Continuous 32- or 33-day chamber exposure of ponderosa pine, white fir, California black oak 12 and canyon live oak to 24-hour average HNO<sub>3</sub> concentrations generally ranging from 10 to 18 13  $\mu g/m^3$  (moderate treatment), or 18 to 42  $\mu g/m^3$  (high treatment), with the average of the highest 14 10% of concentrations generally ranging from 18 to 42  $\mu$ g/m<sup>3</sup> (30-60  $\mu$ g/m<sup>3</sup> peak), or 89 to 155 15  $\mu g/m^3$  (95-160  $\mu g/m^3$  peak), resulted in damage to foliar surfaces of the 1 to 2-year old plants 16 (ISA, Appendix 3, section 3.4; Padgett et al., 2009). The moderate treatment reflects exposure 17 concentrations observed during some summer periods in the Los Angeles Basin in the mid-18
- 19 1980s, including a high HNO<sub>3</sub> concentration of 33  $ug/m^3$  in August 1986 (Padgett et al., 2009;
- 20 Bytnerowicz and Fenn, 1996), when annual average NO<sub>2</sub> concentrations in the Basin ranged up
- 21 to 0.058 ppm (U.S. EPA, 1987).
- 22 The available evidence for lichens includes a recent laboratory study, involving daily HNO<sub>3</sub> exposures for 18 to 78 days, with daily peaks near 50 ppb ( $\sim$ 75 µg/m<sup>3</sup>) reported decreased 23 photosynthesis, among other effects (ISA, Appendix 6, section 6.2.3.3; Riddell et al., 2012). 24 25 Based on studies extending back to the 1980s, HNO<sub>3</sub> has been suspected to have had an important role in the dramatic declines of lichen communities that occurred in the Los Angeles 26 basin (ISA, Appendix 3, section 3.4; Nash and Sigal, 1999; Riddell et al., 2008; Riddell et al., 27 2012). For example, lichen transplanted from clean air habitats to analogous habitats in the Los 28 29 Angeles basin in 1985-86 were affected in a few weeks by mortality and appreciable accumulation of H+ and NO<sub>3</sub><sup>-</sup>(ISA, Appendix 3, section 3.4; Boonpragob et al., 1989). As 30 described in Appendix 5B, section 5B.4.1, the Los Angeles metropolitan area experienced NO<sub>2</sub> 31 concentrations well in excess of the NO<sub>2</sub> secondary standard during this period. For example, 32 annual average NO<sub>2</sub> concentrations in Los Angeles ranged up to 0.078 ppm in 1979 and 33 remained above the standard level of .053 ppm into the early 1990s (Appendix 5B, section 34 5B.4.1). The magnitude and spatial extent of declines over the last several decades, in both dry 35 deposition of HNO<sub>3</sub> and annual average HNO<sub>3</sub> concentration in this area and nationally, are 36

illustrated in Figures 2-40 and 2-41 above (ISA, Appendix 2, Figure 2-60). As assessed in the 1 2 ISA, the evidence indicates NO<sub>2</sub>, and particularly, HNO<sub>3</sub>, as "the main agent of decline of lichen in the Los Angeles basin" (ISA, Appendix 3, p. 3-15), thus indicating a role for the elevated 3 concentrations of nitrogen oxides documented during the 1970s to 1990s (and likely also 4 occurring earlier). More recent studies indicate variation in eutrophic lichen abundance to be 5 6 associated with variation in N deposition metrics (ISA, Appendix 6, section 6.2.3.3). The extent 7 to which these associations are influenced by residual impacts of historic air quality is unclear.

#### 8 5.1.3 Particulate Matter

The extent to which quantitative information is available for airborne PM concentrations 9 associated with ecological effects varies for the various types of effects. The concentrations at 10 which PM has been reported to affect vegetation (e.g., through effects on leaf surfaces which 11 may affect function, or through effects on gas exchange processes) are generally higher than 12 those associated with conditions meeting the current standards and may be focused on specific 13 particulate chemicals rather than on the mixture of chemicals in PM occurring in ambient air. For 14 example, reduced photosynthesis has been reported for rice plants experiencing fly ash particle 15 deposition of 0.5 to 1.5 g/m<sup>2</sup>-day, which corresponds to loading greater than 1000 kg/ha-yr (ISA, 16 Appendix 15, sections 15.4.3 and 15.4.6). Studies involving ambient air PM have generally 17 18 involved conditions that would not be expected to meet the current secondary standards, e.g., 19 polluted locations in India or Argentina (ISA, Appendix 15, sections 15.4.3 and 15.4.4). Studies in the U.S. have looked at the effects of airborne PM on plant reproduction near roadway 20 21 locations in the U.S. have not reported a relationship between PM concentrations and pollen 22 germination (ISA, Appendix 15, section 15.4.6). In summary, little information is available on 23 welfare effects of airborne PM in exposure conditions likely to meet the current standards, and 24 that which is available does not indicate effects to occur under those conditions.

#### 25

### 5.2 AQUATIC ECOSYSTEM ACIDIFICATION

26 Changes in biogeochemical processes and water chemistry caused by deposition of nitrogen and sulfur to surface waters and their watersheds have been well characterized for 27 decades and have ramifications for biological functioning of freshwater ecosystems, as 28 29 summarized in section 4.2.1.1 above (ISA, Appendix 8, section 8.1). This acidifying deposition 30 infiltrates both terrestrial and aquatic systems and may result in changes to soils and water that are harmful to biota. These changes are dependent on a number of factors that influence the 31 32 sensitivity of a system to acidification including weathering rates, bedrock composition, vegetation and microbial processes, physical and chemical characteristics of soils and hydrology. 33

The quantitative assessment of aquatic acidification risk performed for this review is 1 2 based on established modeling approaches, extensive databases of site-specific water quality measurements and a commonly recognized indicator of acidification risk, ANC. Key aspects of 3 this assessment and its results are summarized in the following subsections, with details provided 4 in Appendix 5A. Section 5.2.1 provides background information on the evidence supporting the 5 6 use of ANC as an indicator of acidification risk in the assessment. The conceptual model and the 7 analysis approach are summarized in section 5.2.2. Results for analyses at three scales are presented in section 5.2.3 and a characterization of the analysis uncertainty is summarized in 8 section 5.2.4. Overall findings are summarized in section 5.2.5. 9

10 5.2.1 Role of ANC as Acidification Indicator

Several measures of surface water chemistry are commonly used in assessments of 11 aquatic acidification. These include surface water base cations, pH, inorganic Al and ANC (ISA, 12 Table IS-3). Accordingly, risk to aquatic systems from acidifying deposition can be assessed as a 13 change in specific water quality metrics as a result of nitrogen and/or sulfur deposition. Changes 14 in surface water chemistry reflect the influence of acidic inputs from precipitation, gases, and 15 particles, as well as local geology and soil conditions. As described in section 4.2.1.1.2 above, 16 surface water chemical factors such as pH, Ca<sup>2+</sup>, ANC, ionic metals concentrations, and 17 dissolved organic carbon (DOC) are affected by acid deposition and can profoundly affect the 18 19 structure and function of biological communities in lakes and streams (ISA, Section 8.3). The 20 most widely used measure of surface water acidification, and subsequent recovery under scenarios with lower acidifying deposition, is ANC. 21 22 As summarized in section 4.2.1.1.2 above, the evidence of effects on biota from aquatic

acidification indicates a range of severity with varying ANC levels. The evidence relates to biota
ranging from phytoplankton and invertebrates to fish communities. For example, a review by

Lacoul et al. (2011) of aquatic acidification effects on aquatic organisms in Atlantic Canada

observed that the greatest differences in phytoplankton species richness occurred across a pH

27 range of 4.7 to 5.5 (ANC range of 0 to 20  $\mu$ eq/L), just below the range (pH 5.5 to 6.5) where

bicarbonate becomes rapidly depleted in the water (ISA, Appendix 8, section 8.3.1.1). Under

29 acidifying conditions, these phytoplankton communities shifted from dominance by

- 30 chrysophytes, other flagellates, and diatoms to dominance by larger dinoflagellates. In benthic
- 31 invertebrates residing in sediments of acidic streams, Al concentration is a key influence on the
- 32 presence of sensitive species. Studies of macroinvertebrate species have reported reduced species
- richness at lower pH, with the most sensitive group, mayflies, absent at the lowest levels. Values

- 1 of pH below 5 (which may correspond to ANC levels below 0  $\mu$ eq/L)<sup>2</sup> were associated with the
- 2 virtual elimination of all acid sensitive mayfly and stonefly species over the period from 1937-42
- to 1984-85 in two streams in Ontario (Baker and Christensen, 1991). In a more recent study,
- 4 Baldigo, et al., (2009) showed macroinvertebrate assemblages in the southwestern Adirondack
- 5 Mountains were severely impacted at pH <5.1, moderately impacted at pH from 5.1 to 5.7,
- 6 slightly impacted at pH from 5.7 to 6.4 and usually unaffected above pH 6.4 (Figure 5-1). In
- 7 Atlantic Canada, Lacoul et al. (2011) found the median pH for sensitive invertebrate species
- 8 occurrence was between 5.2 and 6.1 (ANC of 10 and 80 µeq/L), below which such species
- 9 tended to be absent. For example, some benthic macroinvertebrates, including several species of
- 10 mayfly and some gastropods are intolerant of acid conditions and only occur at pH  $\geq$ 5.5 (ANC
- 11 20  $\mu$ eq/L) and  $\geq$ 6, (ANC 50  $\mu$ eq/L) respectively (ISA, Section 8.3.3).
- 12



Figure 5-1. Total macroinvertebrate species richness as a function of pH in 36 streams in
 western Adirondack Mountains of New York, 2003-2005. From Baldigo et al.
 (2009); see ISA, Appendix 8, section 8.3.3 and p. 8-12.

Responses among fish species and life stages to changes in ANC, pH and Al in surface
waters are variable (ISA, Appendix 8, section 8.3.6). Early life stages such as larvae and smolts
are more sensitive to acidic conditions than the young-of-the-year, yearlings, and adults (Baker,
et al., 1990; Johnson et al., 1987; Baker and Schofield). Some species and life stages experienced
significant mortality in bioassays at relatively high pH ((e.g., pH 6.0–6.5; ANC 50-100 µeq/L for

<sup>&</sup>lt;sup>2</sup> pH and ANC were related to one another using a generalized relationship base on equilibrium with atmospheric CO<sub>2</sub> concentration (Cole and Prairie, 2010)

- 1 eggs and fry of striped bass and fathead minnow) (McCormick et al., 1989; Buckler et al.,
- 2 1987)), whereas other species were able to survive at quite low pH without adverse effects.
- 3 Many minnows and dace (Cyprinidae) are highly sensitive to acidity, but some common game
- 4 species such as brook trout, largemouth bass, and smallmouth bass are less sensitive (threshold
- 5 effects at pH <5.0 to near 5.5; ANC 20 and 50 μeq/L). A study by Neff et al. (2008), investigated
- 6 the effects of two acid runoff episodes in the Great Smoke Mountains National Park on native
- 7 brook trout using an in-situ bioassay. The resulting whole-body sodium concentrations before
- 8 and after the episodes showed negative impacts on physiology. More specifically, the reduction
- 9 in whole-body sodium when stream pH dropped below 5.1 (ANC 0  $\mu$ eq/L) indicated that the
- trout had lost the ability to ionoregulate (ISA, Appendix 8, section 8.3.6.1). See Figure 5-2 for
- 11 fish species sensitivity based on observations from field studies with supporting bioassays.



Figure 5-2. Critical aquatic pH range for fish species. Notes: Baker and Christensen
(1991) generally defined bioassay thresholds as statistically significant
increases in mortality or by survival rates less than 50% of survival rates in
control waters. For field surveys, values reported represent pH levels
consistently associated with population absence or loss. Source: Fenn et al.
(2011) based on Baker and Christensen (1991). (ISA, Appendix 8, Figure 8-3)

As noted in the ISA, "[a]cross the eastern U.S., brook trout are often selected as a 1 2 biological indicator of aquatic acidification because they are native to many eastern surface 3 waters and because residents place substantial recreational and aesthetic value on this species" (ISA, Appendix 8, p. 8-26). Compared to other fish species in Appalachian streams, this species 4 is relatively pH sensitive. For example, "[in many Appalachian mountain streams that have been 5 6 acidified by acidic deposition, brook trout is the last fish species to disappear; it is generally lost 7 at pH near 5.0 (MacAvoy and Bulger, 1995), which usually corresponds in these streams with ANC near 0 µeq/L (Sullivan et al., 2003)" (ISA, Appendix 8, p. 8-21). 8

9 As described in section 4.2.1 above episodic acidification during storm events can pose risks in low ANC streams. For example, streams with ANC around 20 µeq/L or less at base flow 10 may be considered vulnerable to episodic acidification events that could reduce pH and ANC to 11 levels potentially harmful to brook trout and other species. Streams with suitable habitat and 12 annual average ANC greater than about 50 µeq/L are often considered suitable for brook trout in 13 14 southeastern U.S. streams and reproducing brook trout populations are expected (Bulger et al., 15 2000). Streams of this type "provide sufficient buffering capacity to prevent acidification from eliminating this species and there is reduced likelihood of lethal storm-induced acidic episodes" 16 (ISA, Appendix 8, p. 8-26). Results of a study by Andrén and Rydin (2012) suggested a 17 threshold less than 20 ug/L Al and pH higher than 5.0 for healthy brown trout populations by 18 exposing yearling trout to a pH and inorganic Al gradient in humic streams in Scandinavia (ISA, 19 20 Appendix 8, section 8.3.6.2). Another recently available study that investigated the effects of episodic pH shifts fluctuations in waterbodies of eastern Maine reported that episodes resulting 21 22 in pH dropping below 5.9 (ANC of  $\sim$ 50 µeq/L) have the potential for harmful physiological effects to Atlantic salmon smolts if coinciding with the smolt migration in eastern Maine rivers 23 24 (Liebich et al., 2011; ISA, Appendix 8, section 8.3.6.2).

25 Investigations of waterbody recovery from historic deposition have reported on episodic 26 acidification associated with the high S absorption remaining in watershed soils. For example, monitoring data in the Great Smoky Mountains National Park indicated that while the majority 27 of  $SO_4^{2-}$  entering the study watershed was retained,  $SO_4^{2-}$  in wet deposition moved more directly 28 29 and rapidly to streams during large precipitation events, contributing to episodic acidification of receiving streams and posing increased risk to biota (ISA, Appendix 7, section 7.1.5.1.4). High 30 flow episodes in historically impacted watersheds of the Appalachians have been reported to 31 appreciably reduce stream ANC (Lawrence et al., 2015). 32

There is often a positive relationship between pH or ANC and number of fish species, at least for pH values between about 5.0 and 6.5, or ANC values between about 0 and 50 to 100  $\mu$ eq/L (Cosby et al., 2006; Sullivan et al., 2006; Bulger et al., 1999). This is because energy cost in maintaining physiological homeostasis, growth, and reproduction is high at low ANC levels

- 1 (Schreck, 1982; Wedemeyer et al., 1990). As noted in section 4.2.1.1.2 above, surveys in the
- 2 heavily impacted Adirondack mountains found that lakes and streams having an annual average
- 3 ANC  $< 0 \mu eq/L$  and pH near or below 5.0 generally support few or no fish species to no fish at
- 4 all, as illustrated in Figure 5-3 below (Sullivan et al., 2006; ISA, Appendix 8, section 8.3.6.3.



Figure 5-3. Number of fish species per lake *versus* acidity status, expressed as ANC, for
 Adirondack lakes. Notes: The data are presented as the mean (filled circles) of
 species richness within 10 μeq/L ANC categories, based on data collected by
 the Adirondacks Lakes Survey Corporation. Source: Modified from Sullivan
 et al. (2006) (ISA, Appendix 8, Figure 8-4).

The data presented in Figure 5-3 above suggest that there could be a loss of fish species with decreases in ANC below a threshold of approximately 50 to 100 µeq/L for lakes (Sullivan et Could be a loss of fish species)

13 al., 2006). For streams in Shenandoah National Park, a statistically robust relationship between

14 ANC and fish species richness was also documented by Bulger et al., (2000). However,

15 interpretation of species richness relationship with ANC can be difficult and misleading, because

16 more species tend to occur in larger lakes and streams as compared with smaller ones,

17 irrespective of acidity (Sullivan et al., 2006) because of increased aquatic habitat complexity in

18 larger lakes and streams (Sullivan et al., 2003; ISA, Appendix 8, section 8.3.6.3).

19 The key biological/ecological effects on aquatic organisms that have been observed in 20 field studies of different acidification levels. Observations of effects in watersheds impacted by 21 historic acidification can also reflect the influence of episodic high flow events that lower pH

- and ANC appreciably below the baseflow ANC (as described above). Studies described above
- are summarized below in the context of ANC ranges:  $<0, 0-20, 20-50, 50-80, and >80 \mu eq/L$ :

- At ANC levels  $<0 \ \mu$ eg/L, aquatic ecosystems have exhibited low to a near loss of aquatic 1 2 diversity and small population sizes. For example, planktonic and macroinvertebrates communities shift to the most acid tolerant species (Lacoul et al., 2011) and mayflies can 3 be eliminated (Baker and Christensen, 1991). A near to complete loss of fish populations 4 5 can occur, including non-acid sensitive native species such as brook trout (Salvelinus fontinalis), northern pike (Esox lucius), and others (Sullivan et al., 2003, 2006; Bulger et 6 7 al., 2000), which is in most cases attributed to elevated inorganic monomeric Al concentration (Baldigo and Murdoch 1997). At this level, aquatic diversity is at its lowest 8 (Bulger et al. 2000, Baldigo et al. 2009, Sullivan et al. 2006) with only acidophilic 9 species being present. 10
- 11 • In waterbodies with ANC levels between 0 and 20 µeq/L, acidophilic species dominate other species (Matuszek and Beggs, 1988; Driscoll et al., 2001) and diversity is low 12 (Bulger et al. 2000, Baldigo et al. 2009, Sullivan et al. 2006). Plankton and 13 macroinvertebrate populations have been observed to decline, and acid-tolerant species 14 have outnumbered non-acid sensitive species (Liebich et al., 2011). Sensitive species are 15 often absent (e.g., brown trout, common shiner, etc.) while non-sensitive fish species 16 17 populations may be reduced (Bulger et al. 2000). Episodic acidification events (e.g., inflow with ANC <0  $\mu$ eq/L and pH< 5), may have lethal impacts on sensitive lifestages 18 of some biota, including brook trout and other fish species (Matuszek and Beggs, 1988; 19 20 Driscoll et al., 2001).
- Levels of ANC between 20 and 50 µeg/L have been associated with the loss and/or 21 reduction in fitness of aquatic biota that are sensitive to acidification in waterbodies of 22 Adirondacks and Appalachians. Such effects included reduced aquatic diversity (Kretser 23 et al., 1989, Lawrence et al., 2015; Dennis, 1995) with some sensitive species missing 24 (Bulger et al., 2000, Sullivan et al., 2006). In historically impacted watersheds, 25 26 waterbodies with ANC below 50 µeq/L are more vulnerable to increased potential for harm associated with episodic acidification (ISA, Appendix 8, section 8.2). 27 Comparatively, acid tolerant species, such as brook trout may have moderate to healthy 28 29 populations, (Kretser et al., 1989, Lawrence et al., 2015; Dennis, 1995).
- At an ANC between 50 to 80 µeq L-1, the fitness and population size of some sensitive 30 species have been affected in some historically impacted watersheds. Levels of ANC 31 above 50 µeq/L are considered suitable for brook trout and most fish species because 32 buffering capacity is sufficient to prevent the likelihood of lethal episodic acidification 33 events (Driscoll et al. 2001; Baker and Christensen 1991). However, depending on other 34 35 factors, the most sensitive species have been reported to experience a reduction in fitness and/or population size in some waterbodies (e.g., blacknose shiner [Baldigo et al., 2009; 36 Kretser et al., 1989, Lawrence et al., 2015; Dennis, 1995]). Reduced fish species richness 37 has also been reported to be affected in Adirondack streams at ANC 50 (Sullivan et al., 38 2006). 39
- Values of ANC >80 μeq/L have not generally been associated with harmful effects on
   biota (Bulger et al., 1999; Driscoll et al., 2001; Kretser et al., 1989; Sullivan et al., 2006).

#### 1 5.2.2 Conceptual Model and Analysis Approach

The impact of N and/or S deposition on aquatic acidification across the U.S. was 2 3 evaluated in this review by developing analyses using a CL approach with ANC as the 4 acidification indicator. This approach provides a means of assessing risk to a group of lakes, streams, and rivers (i.e., waterbodies) in a given area from various levels of N and/or S 5 deposition. ANC was used as the water quality metric where ANC targets (see description of the 6 5 categories above) were used to correspond to different levels of acidification risk. This 7 approach was used to characterize the risk of acidifying deposition on aquatic acidification 8 9 across the CONUS with a focus on acid sensitive areas. This relationship between acidifying deposition of nitrogen and sulfur; water chemistry 10

- 11 changes reflected by changes in ANC and pH; and waterbody health and biodiversity are the
- 12 basis for the quantitative assessments that were performed in this review and provide the
- 13 foundation for describing the potential impacts from acidification occuring under current
- 14 conditions across the U.S. The following schematic (Figure 5-4) represents the conceptual model
- used in the analyses to link these factors.





17 Figure 5-4. Conceptual Model for Aquatic Acidification Analyses

1 In the analyses described below, waterbody estimates of deposition were compared to

- 2 atmospheric loading (CLs) estimated to support ANC levels equal to each of several targets
- 3 (described in section 5.2.3 below). In general, relatively low CL values (i.e., less than 50
- 4 meq/m2/yr) indicate that the watershed has a limited ability to neutralize the addition of acidic
- 5 anions, and hence, it is susceptible to acidification. The higher the CL value, the greater the
- 6 ability of any given watershed to neutralize the additional acidic anions and protect aquatic life.
- 7 Similarly, for any specific ANC target, lower CL estimates are associated with more acid-
- 8 sensitive waterbodies. Further, given the negative relationship between acidic loading and ANC,
- 9 the CL estimates for any one waterbody are lower for the higher ANC targets.
- 10 Key aspects of the assessments described the subsections below include the spatial scales 11 of assessment (section 5.2.2.1), the chemical indicator (section 5.2.2.2), identification of CL
- estimates for this assessment (section 5.2.2.3) and determining exceedances (section 5.2.2.4), as
- 13 well as sources of waterbody deposition estimates (section 5.2.2.5). Also discussed is the
- 14 approach for interpreting results, including with regard to ecosystems with sensitivity to acidic
- deposition, ecosystems for which factors other than deposition are critical influences on
- 16 waterbody ANC, and systems for which nonzero CL estimates cannot be derived for ANC levels
- 17 of interest (section 5.2.2.6). Results of the assessments are presented in section 5.2.3. The
- 18 characterization of uncertainty is described in section 5.2.4 and key observations are summarized
- in section 5.2.5.

### 20 **5.2.2.1** Spatial Scale

For this review, we developed a multi-scale analysis to assess aquatic acidification at 21 three levels of spatial extent: national, ecoregion, and case study. For this analysis, the national 22 assessment included the CONUS only since there is insufficient data available for Hawaii, 23 24 Alaska, and the territories. The Omernik ecoregion classifications were used for the regional 25 assessments and case studies were selected for areas which were likely to be most impacted and for which sufficient data was available. Further discussion of these spatial scales can be found 26 below. Since acidification of waterbodies is controlled by local factors such as geology, 27 28 hydrology, etc. the aquatic CLs for acidification are unique to the waterbody itself and 29 information about the waterbody, like water quality, is needed to determine its CL. For these 30 reasons, CLs were determined at the waterbody level and then summarized at the national, ecoregion, and case study level. The national assessment is a combined summary of aquatic CLs 31 across the CONUS. 32

It is important to note that aquatic ecosystems across the CONUS exhibit a wide range of sensitivity to acidification because of a host of landscape factors, such as geology, hydrology, soils, catchment scale, and vegetation characteristics that control whether a waterbody will be

- 1 acidified by air pollution deposition. Consequently, variations in ecosystem sensitivity must be
- 2 taken into account in order to characterize sensitive populations of waterbodies and relevant
- 3 regions across the CONUS. The EPA's Omernik Ecoregions classifications were used to define
- 4 ecologically relevant, spatially aggregated, acid sensitive regions across the CONUS in order to
- 5 better characterize the regional difference in the impact of deposition driven aquatic acidification
- 6 (Figure 5-5).



#### 8 Figure 5-5. Omernik Ecoregion II areas with ecoregion III subdivisions

Ecoregions are areas of similarity regarding patterns in vegetation, aquatic, and terrestrial
ecosystem components. Available ecoregion categorization schemes include the EPA's Omernik
classifications (Omernik, 1987). Omernik's ecoregions are categorized using a holistic, "weightof-evidence" approach in which the relative importance of factors may vary from region to
region. The method used to map ecoregions is described in Omernik (1987) and classifies
regions through the analysis of the patterns and the composition of biotic and abiotic phenomena

- 15 that affect or reflect differences in ecosystem quality and integrity. Factors include geology,
- 16 physiography, vegetation, climate, soils, land use, wildlife, and hydrology.

1 Three hierarchical levels were developed to distinguish coarser (more general) and finer

- 2 (more detailed) categorization. Level I is the coarsest level, dividing North America into 12
- 3 ecoregions. At level II, the continent is subdivided into 25 ecoregions. Level III is a further
- 4 subdivision of level II and divides CONUS into 105 ecoregions. Level IV is a subdivision of
- 5 level III, and divides CONUS into 967 ecoregions. For the analyses in this review, we used level
- 6 III ecoregions to give the greatest sensitivity for variation in ecoregion response while allowing
- 7 us to aggregate available water quality data to allow representativeness.

8 The case study scale represents the smallest scale at which we performed our analyses 9 and is intended to give some insight into the local impact of aquatic acidification. Five case 10 study areas across the U.S. were examined. These areas were the Shenandoah National Park, 11 White Mountain National Forest, Voyagers National Park, Sierra National Forest, and Rocky 12 Mountain National Park. These parks and national forest vary in their sensitivity to acidification, 13 but represent high value or protected ecosystems, such as Class 1 areas, wilderness, and national 14 forests.

#### 15 5.2.2.2 Chemical Indicator

16 The chemical indicator of acidification risk used in this assessment is ANC. Selection of ANC provides a way to look most closely at those waterbodies for which deposition was the 17 main source of acidifying input as well as eliminating from consideration those waterbodies for 18 19 which either other sources of acidifying input were significant (for example, runoff) or for which 20 natural conditions were such that those waterbodies would be unable to reach specific ANC threshold. Surface water ANC is also commonly used for estimating CLs for N and S in the U.S 21 as it is more stable and more easily modelled. Additionally, CL estimates generally are linearly 22 associated with ANC target, and, unlike some other indicators, ANC is not influenced by other 23 24 environmental factors such as  $CO_2$  levels in the surface water (ISA, section 7.1.2.5).

25 For the analyses described below, we evaluated CLs for three different ANC thresholds:

26 20  $\mu$ eq/L, 30  $\mu$ eq/L and 50  $\mu$ eq/L. Selection of these target ANC values reflect several

considerations. For example, most aquatic CL studies conducted in the U.S. since 2010 use an

ANC of 20 and/or 50 µeq/L, because 20 µeq/L provides protection for "natural" or "historical"

- range of ANC and 50 µeq/L provides overall ecosystem protection (DuPont et al., 2005;
- 30 McDonnell et. al., 2012, 2014; Sullivan et al., 2012a, 2012b; Lynch et al., 2022; Fakhraei et al.,
- 2014; Lawrence et al., 2015). In the western U.S., lakes and streams vulnerable to deposition
- 32 driven aquatic acidification are often found in the mountains where surface water ANC levels are
- naturally low and typically vary between 0 and 30  $\mu$ eq/L (Williams and Labou 2017, Shaw et al.,
- 2014). For these reasons, previous studies and the National Critical Load Database (NCLD),
- used an ANC threshold of 50  $\mu$ eq/L for the eastern CONUS and 20  $\mu$ eq/L for the western

CONUS (denoted as "50/20" µeq/L). With regard to higher ANC levels, such as 80 µeq/L, it was 1 also recognized that many waterbodies, particularly, in acid sensitive regions of CONUS never 2 3 had an ANC that high and would never reach an ANC that high naturally (Williams and Labou 2017, Shaw et al 2014). Additionally, in conveying its advice in the 2012 review, the CASAC 4 expressed its view that "[1]evels of 50 µeq/L and higher would provide additional protection, but 5 6 the Panel has less confidence in the significance of the incremental benefits as the level increases 7 above 50 µeq/L" (Russell and Samet, 2010; pp. 15-16). For the analyses included below ANC target values of 20, 30 and 50 µeq/L were selected 8 9 for the following reasons: 10 ANC of 20  $\mu$ eq/L : In western high elevation sites, ANC is typically below 50 µeq/L (e.g., median 11 around 30 µeq/L in Sierra Nevada) even though acidifying deposition is low at 12 those sites (Shaw et al., 2014). Accordingly, a target of 20 µeq/L is commonly 13 considered an appropriate target for western sites. 14 ANC levels below 20 µeq/L in sensitive Shenandoah/Adirondack waterbodies are 15 \_ associated with significant/appreciable reduction in fish species (Bulger et al. 16 2000; Sullivan et al. 2006). Thus, ANC of 20 µeq/L is considered a 17 18 minimum/lower bound target for such eastern mountain sites. 19 ANC of 30  $\mu$ eq/L: While ecological effects occur at ANC levels at 30 µeq/L in some sensitive 20 ecosystems (based primarily on studies in Shenandoah/Adirondack waterbodies, 21 22 the degree and nature of those effects are less significant than at levels below 20 23  $\mu eq/L$ . 24 Research in New England, the Adirondacks and Northern Appalachian Plateau \_ 25 indicates ANC of 30-40 µeq/L may protect from spring episodic acidification in those watersheds (Driscoll et al. 2001; Baker and Christensen 1991) 26 ANC of 50 µeq/L 27 28 ANC of 50  $\mu$ eq/L is is commonly cited as a target for eastern sites (DuPont et al., 2005; McDonnell et. al., 2012; McDonnell et. al., 2014; Sullivan et al., 2012a; 29 Sullivan et al., 2012b; Lynch et al., 2022; Fakhraei et al., 2014; Lawrence et al., 30 2015). 31 32 - In 2012 review, ANC values at/above 50 were concluded to provide additional protection although with increasingly greater uncertainty for values at/above 75 33 34 μeq/L (2011 PA, pp. 7-47 to 7-48). 5.2.2.3 Critical Load Estimates Based on ANC 35 Considerable new research on critical loads for acidification is available and both steady 36 37 state and dynamic models have been used to generate ANC based critical loads for much of the U.S. Steady-state CLs are calculated from mass-balance models under assumed or modeled 38 39 equilibrium conditions based in part on water quality measurements. While the models used to

derive steady-state CLs vary in complexity, fundamentally they rely on the calculation of 1 2 elemental mass balances. Dynamic models have also been used to develop CLs. These models simulate soil or water chemistry or biological response to calculate a target within a specified 3 time period, such as by the Year 2100, and they can also be used to calculate a CL comparable to 4 a long-term steady-state CL by applying the model to a date in the distant future. Since the 2008 5 6 ISA, studies utilizing dynamic modeling of CLs has generally been focused on the Adirondacks, 7 Appalachians, and the Rocky Mountains/Sierra Nevada (ISA, Appendix 8, section 8.5.4.1.2.2). 8 Empirical studies have also identified CLs for freshwater systems (ISA, Appendix 8, 9 Table 8-7). For example, in the Sierra Nevada mountains, total acidic deposition of about 74 10 eq/ha/yr was correlated with the decline in ANC observed in Moat Lake between 1920 and 1930 11 (Heard et al., 2014). Baron et al. (2011) estimated CLs to be about 571 eq N/ha/yr in the Northeast and 286 eq N/ha/yr in the Rocky Mountains for NO3<sup>-</sup> concentrations as triggers of 12 episodic acidification. In California, CLs for N deposition in California were estimated based in 13 14 part on changes in NO3<sup>-</sup> leaching in stream water, which can cause or contribute to water acidification (Fenn et al., 2008). Critical loads derived empirically and by the DayCent model for 15 NO3<sup>-</sup> leaching were both 1,214 eq N/ha/yr (ISA, Appendix 8, section 8.6.8). 16 There are several newly available studies using steady-state modeling. Sullivan et al. 17 18 (2012b) and McDonnell et al. (2012) developed an approach for deriving regional estimates of base cation weathering to support steady-state CL estimates for the protection of southern 19 20 Appalachian Mountain streams against acidification. Calculated CL values were low at many 21 locations, suggesting high acidification sensitivity. In the Blue Ridge ecoregion, calculated CL 22 values to maintain stream ANC at 50 µeq/L were less than 500 eq/ha/yr at one-third of the study sites. 23 24 In another model simulation for Appalachian Mountain streams, McDonnell et al. (2014) 25 calculated critical values, including steady-state aquatic CLs to protect streams against 26 acidification. They based the CLs on ANC thresholds of 50–100 µeq/L and nearly one-third of 27 the stream length assessed in the study region had a CL for S deposition below < 500 eq/ha/yr 28 (ISA, Appendix 8, section 8.6.8). 29 In the western U.S., Shaw et al. (2014) used the SSWC model to estimate CLs for 2008 lakes in Class I and II wilderness areas in the Sierra Nevada. For benchmark ANC values of 0, 5, 30 10, and 20 µeq/L, which span the range of minimum ANC values observed in Sierra Nevada 31 lakes, median CLs were estimated to be 217, 186, 157, and 101 eq (S + N)/ha/yr to achieve ANC 32 values of 0, 5, 10, and 20 µeq/L, respectively. The median CL for granitic watersheds based on a 33 34 critical ANC limit of 10 µeq/L was 149 eq/ha/yr (ISA, Appendix 8, section 8.5.4.1.2.1). Aquatic CLs used in this assessment came from the NCLD version 3.2.1 (Lynch et al., 35 2022), and studies identified in the ISA (e.g., Shaw et al., 2014; McDonnell et al., 2014; Sullivan 36

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1 et al., 2012a). The NCLD is comprised of CLs calculated from a host of common models. A

- 2 more detailed description of these models can be found in Appendix 5A. Figure 5-6 below shows
- 3 the unique locations for 14,000+ CLs used in this assessment. Critical loads have been developed
- 4 for waterbodies concentrated in areas that are acid sensitive, primarily, the eastern U.S. and the
- 5 Rocky Mountain and Pacific Northwest regions of the west. Not all waterbodies are sensitive to
- 6 acidification. Small to median size lakes (>200 Ha) and lower order streams tend to be the
- 7 waterbodies that are impacted by deposition driven acidification while rivers are not typically
- 8 impacted. Data in the NCLD is focused on waterbodies that are typically impacted by deposition
- 9 driven acidification. A waterbody is represented as a single CL value. In many cases where more
- 10 than one CL value has been estimated for a waterbody (e.g., via different studies) the CL from
- 11 the most recent study was selected or, when the CL estimates are from publications of the same
- 12 timeframe, they are averaged.

#### 13 5.2.2.4 Critical Load-Based Analysis

In this analysis, we compared waterbody estimates to critical loads based on the three 14 ANC targets. A critical load exceedance was concluded when acidifying deposition estimate was 15 16 greater than the target CL. As well documented in the evidence, deposition of both S and N contributes to acid deposition and associated acidification risk of a waterbody. However, as not 17 all N deposition to a watershed will contribute to acidification, evaluating acidic deposition for N 18 and S together is complex. Nitrogen deposition inputs below what is removed by long-term N 19 20 processes in the soil and waterbody (e.g., N uptake and immobilization) do not contribute to acidification, but the amount above this minimum will likely contribute to acidification. 21 Therefore, if N removal is greater than N deposition, only S deposition will contribute to the 22 acidication and thereby to any potential for exceedance of the acidification CL. The analyses 23 24 performed for this PA avoided this complexity by focusing on S only deposition.

The decision to focus on the S component of acidic deposition was based on the less significant contribution of recent N deposition to acidification (compared to past decades). This was concluded based on the finding for deposition in 2014-2016 and 2018-2020 of very few

- exceedances driven by N. This means that adding N from leaching to the critical load
- 29 exceedances with S doesn't really change the percent of waterbodies exceeding their CL. To
- 30 confirm this assumption, analyses were performed to compare the percentage of CL exceedances
- 31 when both N and S were evaluated versus only S exceedances (see Appendix 5A (Section
- 32 5A.2.1). This analysis supported the assumption being used in this assessment that most of the N
- 33 deposition entering the watersheds under the analyses time periods were retained within the
- 34 watershed and/or converted to gaseous N (e.g., N<sub>2</sub>O, N<sub>2</sub>, etc.). Additionally, there were two
- 35 different methods considered for determining the contribution of N deposition to aquatic

acidification. Those methods and how they are handled in CL exceedance calculations are also 1

2 discussed in Appendix 5A.

3 Critical loads and deposition estimates are uncertain and to have confidence in the exceedance it is important that this uncertainty is factored into the calculation. Based on the CL 4 uncertainty analysis (see Appendix 5A, section 5A-2), on average the magnitude of the 5 uncertainty for aquatic CLs is 4.29 meq S/m<sup>2</sup>-yr or 0.69 Kg S/ha-yr with a confidence interval of 6 7  $\pm 2.15 \text{ meg/m}^2/\text{yr}$  or  $\pm 0.35 \text{ Kg}$  S/ha/yr. For simplicity reasons, a 6.25 meg S/m<sup>2</sup>-yr or 1 Kg S/ha/yr range of uncertainty was used in the exceedance calculation. Within this range, it is 8 9 unclear whether the CL is exceeded. For that reason, an exceedance was concluded when the S deposition estimates were greater than the CLs by a margin of  $3.125 \text{ meq S/m}^2$ -yr or 0.5 Kg10 S/ha/yr. An exceedance was not concluded when the S deposition estimate is below the CL by at 11 least a margin of 3.125 meg S/m<sup>2</sup>-yr or 0.5 Kg S/ha/yr. Estimates of S deposition that are within 12 3.125 meq S/m<sup>2</sup>-yr or 0.5 Kg S/ha/yr of the CL are described for the purpose of our analyses as 13 being "at" the CL. 14

#### 15 **5.2.2.5** Waterbody Deposition Estimates

16 Estimates of waterbody deposition used in this assessment were based on the Total Deposition (TDep) model (https://nadp.slh.wisc.edu/committees/tdep/) (Schwede and Lear, 17 2014). This model is discussed more fully in Section 2.5. Both total N and S deposition were 18 19 estimated at a resolution of a 4km grid cell for each stream reach or lake location. For each 20 waterbody, total N and S deposition was determined for each year from 2000 to 2020 and used to derive three-year averages for five periods: 2001-03, 2006-08, 2010-2012, 2014-16 and 2018-20. 21 22 The extent of critical load exceedances was then calculated for each of these five periods and summarized nationally and by ecoregion III (sections 5.2.3.1 and 5.2.3.2). 23

#### 5.2.2.6 Interpreting Results 24

In order to focus our analyses on those areas which were likely to be impacted by 25 26 acidification and that were also driven primarily by deposition of N and S from ambient air, we needed to look more closely at the ecoregions and their underlying characteristics. We also 27 needed to identify those ecoregions where, for various reasons, target ANC values could not be 28 29 achieved. These factors are discussed fully in Appendix 5A and summarized below. 30 The exceedance analysis was performed in waterbodies in 27 ecoregions (level III).

These ecoregions were selected (as described in Appx 5A, section 5A.1.7) based on 31

consideration of their sensitivity to acidification, and their potential for natural (vs deposition-32

driven) acidity (Figure 5-6). Thirty ecoregions were considered sensitive to acidification. Of 33

34 these 30 ecoregions, three were identified as having natural acidity, based on DOC as an

35 indicator of natural acidity. The acid sensitive ecoregions generally are areas with mountains,

- 1 high elevation terrain or waterbodies in northern latitudes (northern areas of Minnesota,
- 2 Wisconsin, and Michigan; and New England). The northern, non-mountainous regions that are
- 3 sensitive share attributes (e.g., growing season, vegetation, soils, and geology) similar to
- 4 mountainous regions and typically are located in rural areas, often in tracts of designated
- 5 wilderness, park and recreation areas. The three naturally acidic ecoregions, located on eastern
- 6 coastal plain, were excluded from the analyses because of their natural acidity indicated by high
- 7 DOC values: (1) Middle Atlantic Coastal Plain (8.5.1), (2) Southern Coastal Plains (8.5.3), and
- 8 (3) Atlantic Coastal Pine Barrens (8.5.4). A more complete discussion of each ecoregion and its
- 9 sensitivity can be found in Appendix 5A (Table 5A-5).



- 14 Estimates of CL less than zero indicate that no level of acidifying deposition would allow
- those areas to reach a target ANC value. These areas, by and large, are those that due to either
- base cation loss from past deposition or natural conditions would not be able to achieve the target
- 17 ANC values of 20, 30 or 50  $\mu$ eq/L under any deposition scenario. These areas were tracked

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12

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separately from those areas with non-zero CL estimates. A more complete discussion of negative
 CLs and results can be found in Appendix 5A.

#### 3 5.2.3 Estimates for Achieving ANC Targets with Different Deposition Levels

The aquatic acidification assessments developed for this review are intended to estimate the ecological exposure and risk posed to aquatic ecosystems from the acidification effects of S and/or N deposition at varying levels to sensitive regions across the CONUS. They were performed at three spatial scales of differing levels of complexity. The results of these analyses are presented below. Section 5.2.3.1 presents the results of the national scale analyses whereas sections 5.2.3.2 and 5.2.3.3 present the results of the ecoregion scale and case study analyses respectively.

#### 11 5.2.3.1 National Scale Analysis

A total of 13,824 unique waterbodies across the CONUS had calculated CLs. Most of 12 13 those waterbodies had CLs that were less than 18 kg S/ha-yr across all the target ANC levels (Appendix 5A, Table 5A-6). Table 5-1 contains a summary of the percent of waterbodies with 14 15 CL exceedances for S only for annual average deposition in the five 3-year periods for the ANC 16 thresholds for an ANC of 20, 30, 50, and  $50/20 \mu eq/L$ . Note that as discussed above, for the purpose of this analysis we focused on CL>0 and S only. The 50/20 values reflect a threshold 17 ANC of 50 µeq/L in the eastern portion of the U.S. and a target ANC of 20 µeq/L in the west. 18 See discussion above for parameters used in developing this scenario. 19

# 20Table 5-1.Percentage of waterbodies nationally for which annual average S deposition21during the five time periods assessed exceed the waterbody CL for each of the22ANC targets.

ANC (µeq/L)	2018-20	2014-16	2010-12	2006-08	2001-03
20	1%	3%	5%	16%	22%
30	2%	4%	7%	19%	25%
50	4%	6%	11%	24%	28%
50/20	4%	6%	10%	23%	28%

23

The geographic distribution of the waterbodies for which S deposition during the five time periods exceeded CLs for the target ANC values is shown in Figures 5-7 to 5-11. Most exceedances occurred in New England, the Adirondacks, the Appalachian Mountain range (New England to Georgia), the upper Midwest, Florida, and the Sierra Nevada mountains in California as expected. As discussed above, waterbodies in Florida that exceed the CL are likely not related to deposition of S, but instead are related to high levels of natural acidity in these drainage waters. These drainage waters tend to be naturally high in dissolved organic carbon, causing

- 1 these systems to be acidic. Because these are waterbodies that are highly sensitive to
- 2 acidification and likely naturally acidic, they exceed the calculated CL at any deposition amount.
- 3 For these reasons, these sites have been removed from the assessment. For more information on
- 4 these areas see Appendix 5A, section 5A.2.1.



5 6 Fi

7

Figure 5-7. Waterbodies for which annual average S only deposition for 2001-03 exceed CLs for ANC thresholds: a. 20, b. 30, c. 50, d, 50/20 µeq/L.



Figure 5-8. Waterbodies for which annual average S only deposition for 2006-08 exceed
 CLs for ANC thresholds: a. 20, b. 30, c. 50, d, 50/20 μeq/L.



1

Figure 5-9. Waterbodies for which annual average S only deposition for 2010-12 exceed CLs for ANC thresholds: a. 20, b. 30, c. 50, d, 50/20 µeq/L.



Figure 5-10. Waterbodies for which annual average S only deposition for 2014-16 exceed CLs for ANC thresholds: a. 20, b. 30, c. 50, d, 50/20 µeq/L.



Figure 5-11. Waterbodies for which annual average S only deposition for 2018-20 exceed
 CLs for ANC thresholds: a. 20, b. 30, c. 50, d, 50/20 μeq/L.

The results of the national scale analyses show a significant reduction in exceedances over time as sulfur deposition has decreased (see 2.3.1 for deposition trends). It can also tell us about the levels of deposition that occurred in those time periods and provide the foundation for the additional analyses below to look at what impacts might be expected under different geographic scales and deposition scenarios.

#### 9 5.2.3.2 Ecoregion Analyses

1

Ecoregion-level analyses, summarized below, provide further characterization of both spatial variability of acid sensitive waterbodies across the U.S. and the extent of deposition driven acidification impacts. Since the acidification of waterbodies is controlled by local factors such as geology and hydrology, aquatic CLs for acidification are unique to the waterbody itself and information about the waterbody, like water quality, is needed to determine its critical load. Unfortunately, not all waterbodies within an ecoregion have sufficient data to calculate a CL. This is the case for many ecoregion IIIs (from this point on ecoregion, at the level III,

specification, will be referred to as ecoregions), except for ones that historically are known to be

- 1 in acid sensitive areas since acid sensitive areas typically have been heavily sampled, hence,
- 2 contain many CLs (see Figure 5-12). These areas tend to be in the eastern CONUS in such
- 3 ecoregions as Central Appalachian, Atlantic Maritime Highlands, and the Blue Ridge. Areas in
- 4 the Rockies and Sierra Nevada also have been sampled extensively and contain many CLs. More
- 5 CLs in an ecoregion helps to capture the spatial variability of acid sensitive areas across the
- 6 landscape and provide a more accurate measurement of the impact of deposition driven
- 7 acidification. Ecoregions with few CLs, however, fail to capture the spatial variability of acid
- 8 sensitive areas, which in turn reduces the accuracy of the percentile CL value and limits our
- 9 confidence in the estimated percent of exceedances. For this reason, ecoregions containing
- 10 greater than 50 CLs were the focus of this analysis.
- 11 For the CONUS there are a total of 105 ecoregions of which 25 met the criteria of 50 or
- more CLs (and excluding the three naturally acidic eastern ecoregions), yielding 18 in the east
- 13 and 7 in the west. The Northern Appalachian and Atlantic Maritime Highlands ecoregion had the
- 14 most CLs at 2,851 (see Appendix 5A, Table 5A-10).



16 Figure 5-12. Locations of aquatic critical loads mapped across Ecoregions III.

For each of the 25 ecoregions meeting the CL criteria for this analysis, median annual average S deposition was determined for each 3-year period using a GIS zonal statistic. The minimum to maximum range for median S deposition in these ecoregions was 0.90-18.08 Kg

20 S/ha-yr for 2001-2003 and 0.54-3.64 Kg S/ha-yr for 2018 – 2020 (Table 5-2). Deposition for the

- 1 18 eastern ecoregions had a median value of 11.0 Kg S/ha-yr in 2001-03 and 1.9 Kg S/ha-yr in
- 2 2018-20 (Table 5A-25). Total S deposition for the seven western ecoregions was lower in each
- 3 3-year period, ranging from a median of 1.14 Kg S/ha-yr in 2001-03 to 0.84 Kg S/ha-yr in
- 4 20180-20. For the period 2001-2003, 17 of the 25 ecoregions had a median total S deposition
- 5 over 10 Kg S/ha-yr while there were none over 10 Kg S/ha-yr in the period 2018-2020.
- 6 Ecoregions with the highest median total S deposition were Western Allegheny Plateau, Erie
- 7 Drift Plain, North Central Appalachians, Central Appalachians, Northern Piedmont, Eastern
- 8 Corn Belt Plains, Southwestern Appalachians, and Ridge and Valley, all in the Mid-Atlantic
- 9 region of the eastern U.S (see Appendix 5A, Table 5A-14).

## 10Table 5-2.Min, max, and median total S deposition for the 25 ecoregions included in the<br/>analyses. Deposition values were determined by a zonal statistic for each<br/>ecoregion.12ecoregion.

	Total Sulfur Deposition (Kg S/ha-yr)													
	2001-03	2006-08	2010-12	2014-16	2018-20									
Minimum	0.90	0.98	0.83	0.79	0.54									
Maximum	18.08	15.05	7.24	4.70	3.64									
Median	9.57	8.05	4.34	2.62	1.87									

13

For waterbodies in the 25 ecoregions, this ecoregion-scale analysis compared the 14 ecoregion S deposition estimates in each of the five periods of deposition to the waterbody-15 specific CLs and evaluate the exceedances per ecoregion (Appendix 5A, section 5A.2.2.1). There 16 were no exceedances of any of the ANC thresholds in the west, so we focus here on the eastern 17 ecoregions. We summarize these results for the 18 eastern ecoregions below, in terms of number 18 19 and percentage of waterbodies per ecoregion with Cl exceedances in every ecoregion-time period combination, using ecoregion deposition estimates as the organizing parameter. For example, 20 Table 5.3 presents the CL exceedance results of the ecoregion level analyses for the three ANC 21 22 target levels, summarized by ecoregion median annual average S deposition (regardless of the 3year period in which it occurred). For each kg S/ha-yr, Table 5.3 presents the number of 23 24 ecoregion-time period combinations with 10, 15, 20, 25 and 30% of waterbodies exceeding their CL for the specified ANC target level. 25 For example, among the eastern ecoregion-time period combinations with S deposition at 26 27 or below 2 Kg S/ha-yr across ecoregions and deposition periods there are no ecoregions that have more than 10% of their waterbodies exceeding their CLs for an of the three ANC targets 28 29 (Table 5-3). In contrast, for annual average S deposition at or below 10 Kg S/ha-yr, there are 22 30 of the 90 ecoregion-time period combinations with more than 10% of their waterbodies exceeding their CLs for an ANC of 50  $\mu$ eq/L, one of which had with more than 30% of its 31

- 1 waterbodies exceeding their CLs. The lowest annual average S deposition level associated with
- 2 any ecoregion-time period combinations having more than 30% of waterbodies exceeding their
- 3 CLs was 10 Kg S/ha-yr, for which one ecoregion-time periods had more than 30% of the
- 4 waterbodies exceeding their CLs for all three ANC targets.

S Deposition (Kg/ha-yr):	No. of Eastern Ecoregion- Time Periods	Numl wate	Number of eastern ecoregion-time periods with more than specified percent of waterbodies exceeding their CLs											Number of western ecoregion-time periods with more than 10% waterbodies exceeding CL for ANC target of 20, 30 or 50 µeq/L										
		10%	15%	20%	25%	30%	10%	15%	20%	25%	30%	10%	15%	20%	25%	30%	S Deposition	No. ecoregion	>10%					
		ANC target of 20 µeq/L					ANC target of 30 µeq/L					ANC target of 50 µeq/L					(kg/Ha-yr)	-time periods						
<u>&lt;</u> 2	10	0	0	0	0	0	0	0	0	0	0	0	0	0	0	0	<u>&lt;</u> 2	52	0					
<u>&lt;</u> 3	29	0	0	0	0	0	0	0	0	0	0	1	0	0	0	0	<u>&lt;</u> 3	55	0					
<u>&lt;</u> 5	51	2	1	0	0	0	4	1	0	0	0	9	3	2	1	0	None of the 75 western ecoregion- time periods in analysis had ecoregic							
<u>&lt;</u> 6	59	4	1	0	0	0	7	1	0	0	0	13	4	2	1	0								
<u>&lt;</u> 7	63	5	1	0	0	0	8	2	0	0	0	14	5	3	1	0	S deposition estimates above 3 kg							
<u>&lt;</u> 8	67	9	4	0	0	0	12	6	1	0	0	18	9	5	3	0	S/na-yi							
<u>&lt;</u> 9	69	9	4	0	0	0	13	6	1	0	0	19	9	5	3	0	]							
<u>&lt;</u> 10	73	11	6	1	1	1	16	8	2	1	1	22	11	6	4	1	]							
<u>&lt;</u> 11	76	13	7	2	1	1	18	9	3	1	1	24	13	7	4	1								
<u>&lt;</u> 12	79	15	9	4	3	2	21	11	5	3	3	27	15	9	6	3								
<u>&lt;</u> 13	81	16	10	4	3	2	22	12	5	3	3	28	16	10	6	3	]							
<u>&lt;</u> 14	84	19	12	6	4	3	25	14	7	5	4	31	18	12	8	5	]							
<u>&lt;</u> 15	86	21	14	8	6	4	27	16	9	7	6	33	20	14	10	7								
<u>&lt;</u> 16	88	22	15	9	7	5	28	17	10	8	7	34	21	15	11	8								
<u>&lt;</u> 17	88	22	15	9	7	5	28	17	10	8	7	34	21	15	11	8								
<u>&lt;</u> 18	90	24	17	11	9	7	30	19	12	10	9	36	23	17	13	10	]							

Table 5-3.Number of ecoregion-time period combinations with more than 10, 15, 20, 25, and 30% of waterbodies exceeding<br/>their CLs for three ANC targets as a function of ecoregion-level estimates of annual average S deposition.

- We also considered these ecoregion-scale results from the perspective of the extent to 1 which waterbodies within the eastern ecoregions were estimated to achieve the various ANC 2 3 targets across the S deposition levels for the 18 ecoregions and five time periods. This can be 4 considered the inverse of the presentation in Table 5-3 above, using percentages instead of absolute counts in the presentation. For example, rather than the number of ecoregion-time 5 6 periods, with a particular S deposition estimate, that have more than 10% of waterbodies exceeding their CLs for an ANC target of 20 µeq/L, Figure 5-13 presents the percentage of 7 ecoregion-time periods that have less than or equal to 10% of waterbodies exceeding their CLs 8 for each of the three ANC levels (20, 30 and 50 µeq/L). The same dataset is presented in Table 9 5-4 with the bins for percentage of waterbodies exceeding their CLs (>10, 15, 20, 25 and 30%) 10 flipped to be described as percentage of waterbodies that are at or below their CLs (i.e., can 11 achieve the ANC target). The complete results can be found in Appendix 5A, Section 5A.2.2. 12
- 13





Figure 5-13. Percentage of ecoregion-time period combinations with less than or equal to
 10, 15, 20, 25, and 30% of waterbodies exceeding their CLs for ANC of 20
 (top), 30 (middle) and 50 μeq/L (bottom) for 18 eastern ecoregions.

1	As noted above, Table 5-4 presents the same dataset with the bins for percentage of
2	waterbodies exceeding their CLs (>10, 15, 20, 25 and 30%) flipped to be described as percentage
3	of waterbodies that are expected to achieve an ANC at/above the specified target. Overall, the S
4	deposition levels in the 18 eastern ecoregions analyzed includes a range from below 2 up to
5	nearly 18 kg/ha-yr. Across all 90 eastern ecoregion-time period combinations (including S
6	deposition estimates up to near 18 kg/ha-yr), 73% of the combinations had at least 90% of
7	waterbodies per ecoregion estimated to achieve ANC at or above 20 µeq/L, and 60% had at least
8	90% of the waterbodies estimated to achieve ANC at or above 50 $\mu$ eq/L. Less than half of the
9	eastern ecoregion-time period combinations (and all of the western combinations) had an S
10	deposition estimate below 4 kg/ha-yr. Ninety percent of the eastern combinations were at or
11	below 13 kg/ha-yr. The results by annual average S deposition bin are summarized below for the
12	bins from 13 kg/ha-yr down to 5 kg/ha-yr (the bin that includes at least half of this dataset):
13 14 15	<ul> <li>For S deposition estimates at or below 13 kg/ha-yr, at least 90% of waterbodies per ecoregion were estimated to achieve an ANC at or above 20, 30 and 50 µeq/L in 80, 73 and 65% of all ecoregion-time period combinations, respectively.</li> </ul>
16 17 18	<ul> <li>For S deposition at or below 11 kg/h-yr, at least 90% of all waterbodies per ecoregion were estimated to achieve ANC at or above 20, 30 and 50 μeq/L in 83, 77 and 68% of all ecoregion-time period combinations, respectively.</li> </ul>
19 20 21	• For S deposition at or below 9 kg/h-yr, at least 90% of all waterbodies per ecoregion were estimated to achieve ANC at or above 20, 30 and 50 µeq/L in 87, 81 and 72% of combinations, respectively.
22 23 24	<ul> <li>At least 80%, 75% and 70% of waterbodies per ecoregion were estimated to achieve ANC at or above 20, 30 and 50 µeq/L, respectively, in all ecoregion-time period combinations.</li> </ul>
25 26 27	• For S deposition at or below 7 kg/h-yr, at least 90% of waterbodies per ecoregion were estimated to achieve ANC at or above 20, 30 and 50 µeq/L in 92, 87 and 78% of combinations, respectively.
28 29 30	<ul> <li>At least 80, 80 and 70% of waterbodies per ecoregion were estimated to achieve ANC at or above 20, 30 and 50 µeq/L, respectively, in all ecoregion-time period combinations.</li> </ul>
31 32 33	<ul> <li>For S deposition at or below 5 kg/h-yr, at least 90% of all waterbodies per region were estimated to achieve ANC at or above 20, 30 and 50 µeq/L in 96, 92 and 82% of combinations, respectively.</li> </ul>
34 35 36	<ul> <li>At least 80%, 80% and 70% of waterbodies per ecoregion were estimated to achieve ANC at or above 20, 30 and 50 μeq/L, respectively, in all ecoregion-time period combinations.</li> </ul>
37 38 39	<ul> <li>For S deposition at or below 4 kg/h-yr, at least 90% of all waterbodies per region were estimated to achieve ANC at or above 20 in all 41 ecoregion-time period combinations for that deposition bin, and to achieve ANC at or above 30 and 50 µeq/L in 95 and 97%</li> </ul>

- of those combinations, respectively. The number of ecoregion-time period combinations
   in this deposition bin is less than half the full dataset for the 18 eastern ecoregions.
- For the 75 western-time period combinations, all of which had an S deposition estimate
   below 4 kg/ha-yr, at least 90% of waterbodies per ecoregion were estimated to achieve an
   ANC at or above 50 μg/L.

Total Sulfur	No. of	% Waterbodies per ecoregion-time period meeting specified ANC target															
Deposition (Kg S/ha-vr)	Ecoregi	90%	85%	80%	75%	70%	90%	85%	80%	75%	70%	90%	85%	80%	75%	70%	
at/below:	Periods		ANC ta	rget of 2	0 µeq/L			ANC to	arget of 30	0 µeq/L		ANC target of 50 µeq/L					
2	10	100%	100%	100%	100%	100%	100%	100%	100%	100%	100%	100%	100%	100%	100%	100%	
3	29	100%	100%	100%	100%	100%	100%	100%	100%	100%	100%	97%	100%	100%	100%	100%	
4	41	100%	100%	100%	100%	100%	95%	100%	100%	100%	100%	93%	98%	100%	100%	100%	
5	51	96%	98%	100%	100%	100%	92%	98%	100%	100%	100%	82%	94%	96%	98%	100%	
6	59	93%	98%	100%	100%	100%	88%	98%	100%	100%	100%	78%	93%	97%	98%	100%	
7	63	92%	98%	100%	100%	100%	87%	97%	100%	100%	100%	78%	92%	95%	98%	100%	
8	67	87%	94%	100%	100%	100%	82%	91%	99%	100%	100%	73%	87%	93%	96%	100%	
9	69	87%	94%	100%	100%	100%	81%	91%	99%	100%	100%	72%	87%	93%	96%	100%	
10	73	85%	92%	99%	99%	99%	78%	89%	97%	99%	99%	70%	85%	92%	95%	99%	
11	76	83%	91%	97%	99%	99%	76%	88%	96%	99%	99%	68%	83%	91%	95%	99%	
12	79	81%	89%	95%	96%	97%	73%	86%	94%	96%	96%	66%	81%	89%	92%	96%	
13	81	80%	88%	95%	96%	98%	73%	85%	94%	96%	96%	65%	80%	88%	93%	96%	
14	84	77%	86%	93%	95%	96%	70%	83%	92%	94%	95%	63%	79%	86%	90%	94%	
15	86	76%	84%	91%	93%	95%	69%	81%	90%	92%	93%	62%	77%	84%	88%	92%	
16	88	75%	83%	90%	92%	94%	68%	81%	89%	91%	92%	61%	76%	83%	88%	91%	
17	88	75%	83%	90%	92%	94%	68%	81%	89%	91%	92%	61%	76%	83%	88%	91%	
18	90	73%	81%	88%	90%	92%	67%	79%	87%	89%	90%	60%	74%	81%	86%	89%	

Table 5-4. Percentage of ecoregion-time periods combinations with at least 90, 85, 80, 75 and 70% of waterbodies estimated to achieve an ANC at/above the ANC targets of 20, 30 and 50 µeq/L as a function of annual average S deposition for 18 eastern ecoregions (90 ecoregion-time period combinations).

To further describe how these results compare to recent conditions, we looked at sulfur deposition for eastern ecoregions under the two most recent time periods, 20014-2016 and 2018-2020 and the critical load exceedances that would be expected for the targeted ANC levels of 50, 30 and 20 µeq/L. As would be expected, given deposition trends, there were fewer exceedances in the most recent time periods. Figures 5-14 and 5-15 show the results of these analyses.



Figure 5-14. Percentage of waterbodies in each of the 18 eastern ecoregions exceeding their
 CL for ANC values of 20, 30 and 50 μeq/L, based on annual average S
 deposition for 2014-2016.



11

7

Figure 5-15. Percentage of waterbodies in each of the 18 eastern ecoregions exceeding their
 CL for ANC values of 20, 30 and 50 μeq/L, based on annual average S
 deposition for 2018-2020.
- 1 Three or fewer of 18 eastern ecoregions have more than 10% of their waterbodies
- 2 exceeding the Cl for all of the target ANC values for either time period. The median sulfur
- deposition for eastern ecoregions included in the analyses for the 2014-2016 time period was 3.0
- 4 and for the 2018 2020 time period was approximately 1.9 kg/ha/yr. Figure 5-16 through 5-18
- 5 show the eastern ecoregions with exceedances of target critical loads under the two most recent
- 6 time periods. Figure 5-19 shows the ecoregions with exceedances for the entire U.S. for the most
- 7 recent time periods using an ANC target of 50  $\mu$ eq/L for the east and 20 for the west.

2018 - 2020 Sulfur Deposition Ecoregion Exceedances



2014 - 2016 Sulfur Deposition Ecoregion Exceedances



2 3

Figure 5-16. Map of critical load exceedances for S only deposition from 2018-20 (top) and 2014-16 (bottom) for ANC threshold of 20 µeq/L.



2018 - 2020 Sulfur Deposition Ecoregion Exceedances

2014 - 2016 Sulfur Deposition Ecoregion Exceedances



Figure 5-17. Map of critical load exceedances for S only deposition from 2018-20 (top) and
 2014-16 (bottom) for an ANC threshold of 30 μeq/L.



2018 - 2020 Sulfur Deposition Ecoregion Exceedances

2014 - 2016 Sulfur Deposition Ecoregion Exceedances



Figure 5-18. Map of critical load exceedances for S only deposition from 2018-20 (top) and 2014-16 (bottom) for an ANC threshold of 50 μeq/L.



2018 - 2020 Sulfur Deposition Ecoregion Exceedances

2014 - 2016 Sulfur Deposition Ecoregion Exceedances



ure 5-19. Map of critical load exceedances for S only deposition from 2018-20 (top) and 2014-16 (bottom) for an ANC threshold of 50 µeq/L for East and 20 µeq/L for the West.

#### 1 5.2.3.3 Case Study Analyses

- 2 The case study areas are geographically diverse acid sensitive areas across the CONUS
- 3 that have sufficient data to complete the quantitative analyses. Five case study areas were
- 4 identified that meet the criteria (Figure 5-20), three in the eastern U.S. (NOMN, SHVA and
- 5 WHMT) and two areas are in the western U.S. (ROMO and SINE). Three of the five areas
- 6 (SHVA, ROMO and SINE) are inclusive of Class I areas. Additional aquatic acidification
- 7 analyses using the case studies can be found in Appendix 5A. A total of 524 CLs were found in
- 8 the 5 case study areas, excluding SHVA which had complete coverage (4977 CLs). ROMO,
- 9 SINE, NOMN, and WHMT had 121, 139, 183, and 74 CLs respectively. For this discussion, we
- 10 will refer to analyses that looked at the calculated sulfur deposition values at or below which the
- 11 case study sites would likely be able to attain the target ANC values of 50, 30 and 20  $\mu$ eq/L for
- 12 the eastern case studies and 20  $\mu$ eq/L for the western case studies.



- 13
- Figure 5-20. Location of the case study areas. Northern Minnesota (NOMN), Rocky
   Mountain National Park (ROMO), Shenandoah Valley (SHVA), Sierra
   Nevada Mountains (SINE) and White Mountain National Forest (WHMT).
- 17

# Table 5-5. Annual average S deposition at/below which modeling indicates an ANC of 20, 30 or 50 μeq/L can be achieved in the average, 70% and 90% of waterbodies in each study area.

ANC (µeq/L)	Based on average across all sites in area				Based on 70% of sites achieving				Based on 90% of sites achieving						
	Eastern			Western		Eastern		Western		Eastern			Western		
	N. Minn	White Mtns	Shenan- doah	Rocky Mtn NP	Sierra Nev Mtns	N. Minn	White Mtns	Shenan- doah	Rocky Mtn NP	Sierra Nev Mtns	N. Minn	White Mtns	Shenan- doah	Rocky Mtn NP	Sierra Nev Mtns
	(kg/ha -yr)	(kg/ha- yr)	(kg/ha- yr)	(kg/ha -yr)	(kg/ha- yr)	(kg/ha -yr)	(kg/ha -yr)	(kg/ha- yr)	(kg/ha -yr)	(kg/ha- yr)	(kg/ha -yr)	(kg/ha -yr)	(kg/ha- yr)	(kg/ha -yr)	(kg/ha- yr)
20	11	11	12	9.5	12	5.5	6.9	9.4	5.4	4.1	4.2	4.4	7.1	3.6	1.8
30	10	10	11			5.3	6.1	8.4			3.9	3.3	6.3		
50	10	10	9.4			4.7	4.1	6.3			3.2	0.7	4.1		
Note: Consistent with convention followed in the ecoregion analysis above, CLs are not presented for ANC target values of 30 and 50 μg/L in the west (shading).															

4 The steady-state mass balance modeling results summarized in Table 5-5 indicates the average CL for achieving a target ANC of 20 µeq/L in the five study areas ranges from about 10 5 to 12 kg/ha-yr. For 70 to 90% of sites to achieve an ANC of 20 µeq/L, the estimated CL for S 6 deposition ranges from about 4 to 9 kg/ha-yr. The average CL to achieve an ANC value of 30 7 8 µeq/L ranges from about 10 to 11 kg/ha-yr and for 70-90% of sites to achieve an ANC of 30 µeq/L, the estimated CL for S deposition ranges from about 3 to 8 kg/ha-yr. For an ANC target 9 of 50  $\mu$ eq/L, the average CL for sites in the five case studies ranges from about 7 to 10 kg/ha-yr. 10 For 70 to 90% of the case study sites to achieve a target ANC of 50  $\mu$ eq/L, the estimated CO for 11 S deposition ranges between 3 to 4kg/ha/yr, except for White Mountain, which is extremely 12 13 sensitive. Overall, these findings are slightly lower than the ecoregion scale results.

#### 14 5.2.4 Uncertainty Analyses

15 Models used to estimate CLs, drawn from the NCLD, were derived using a variety of

16 commonly used models, including the steady-state mass-balance model, Steady State Water

17 Chemistry (SSWC) model, and dynamic models such as the Model of Acidification of

18 Groundwater In Catchment (MAGIC) run out to year 2011 or 3000. Key parameters in this

19 modeling include estimates of the catchment-average base-cation supply (i.e., input of base

20 cations from weathering of bedrock and soils and air), runoff, and surface water chemistry.

21 Uncertainty associated with runoff and surface water measurements is not characterized here.

22 The catchment supply of base cations from the weathering of bedrock and soils is the factor that

has the most influence on the CL calculation and has the largest uncertainty (Li and McNulty,

24 2007). For example, the well-established models generally rely on input or simulated values for base

cation weathering (BCw) rate, a parameter the ISA notes to be "one of the most influential yet difficult to

estimate parameters in the calculation of critical acid loads of N and S deposition for protection against

1 terrestrial acidification" (ISA, section IS.14.2.2.1). Obtaining accurate estimates of weathering rates is

- 2 difficult because weathering is a process that occurs over very long periods of time, and the estimates on
- 3 an ecosystem's ability to buffer acid deposition rely on accurate estimates of weathering. Although the
- 4 approach to estimate base-cation supply for the national case study (e.g., F-factor approach) has
- 5 been widely published and analyzed in Canada and Europe, and has been applied in the CONUS

6 (e.g., Dupont et al., 2005 and others), the uncertainty in this estimate is unclear and could be

7 large in some cases. A quantitative uncertainty analysis was completed to evaluate the

8 uncertainty in the CL and exceedance estimation that were used in these analyses (as described

9 further in Appendix 5A, section 5A.3).

Monte Carlo analyses (described in detail in Appendix 5A, section 5A.3.1) were used to 10 describe the 5<sup>th</sup> and 95<sup>th</sup> confidence intervals around the CL for more than 14,000 waterbodies in 11 order to estimate the uncertainty around the CLs. The magnitude of the confidence interval for 12 the CLs was 7.68 meq S/m<sup>2</sup>-yr or 1.3 Kg S/ha/yr. The range based on the 5th to 95th magnitude 13 of the confidence interval was  $0.37-33.2 \text{ meg/m}^2/\text{yr}$  or 0.1-5.3 Kg S/ha/yr giving a confidence 14 level of  $\pm 3.84 \text{ meq/m}^2/\text{yr}$  or  $\pm 0.65 \text{ Kg S/ha/yr}$ . Sixty-one percent of CL values had a low 15 confidence level of less than 3.0325 meq/m<sup>2</sup>/yr or 0.5 Kg S/ha/yr, while 26% had levels greater 16 than 6.25 meq/m<sup>2</sup>/yr or 1.0 Kg S/ha/yr (Appendix 5A, Table 5A-49). Low confidence intervals 17 were associated with CLs determined with long-term water quality data and low variability in 18 runoff measurements. CL values determined by a single water quality measurement and in areas 19 where runoff is variable (e.g., western U.S.) had high uncertainty. Fifty-one ecoregions had 20 sufficient data to calculate the 5<sup>th</sup> to 95<sup>th</sup> percentile (Appendix 5A, Table 5A-50). CLs with the 21 lowest uncertainty occurred in the eastern U.S., particularly along the Appalachian Mountains, 22 upper midwest, and Rockies Mountains (Appendix 5A, Figure 5A-54). Less certain CLs were 23 24 found in the midwest and south and along the CA to WA coast. Most of the CLs in the midwest 25 are based on a single or few water quality measurements while variability in runoff in CA to WA 26 coast account for those high uncertainty values.

27 The magnitude of the error for the N leaching method used in the analyses was estimated by quantifying the uncertainty of the flux of nitrate  $(NO_3)$  to a given lake or stream. Water 28 quality data for the past 28 years from the EPA's Long-term Monitoring (LTM) program was 29 used to assess the uncertainty of the influx of nitrate (NO<sub>3</sub><sup>-</sup>). The results of his uncertainty 30 analysis are summarized in (Appendix 5A, Table 5A-51) by region and time period. Overall, 31 32 nitrate flux varied between regions with Adirondacks lakes having the highest annual fluxes and New England Lakes with the lowest fluxes. While a comprehensive analysis of uncertainty has 33 not been completed for these data prior to the analysis included in this review, expert judgment 34 35 suggested the uncertainty for combined N and S CLs is on average about  $\pm 0.5$  kg/ha-yr (3.125 meq/m<sup>2</sup>/yr), which is consistent with the range of  $\pm 2.30$  to 3.77 meq/m<sup>2</sup>-yr determined from this 36

May 2023

analysis. Given this consistency, an uncertainty of ±3.125 meq/m<sup>2</sup>-yr was applied to the critical
 load exceedances for the national, ecoregion, and case studies assessments.

Critical loads used in the national assessment analysis used different methods than those 3 in the ecoregion and case study analyses (see Appendix 5A, section 5A.1.5). To understand 4 differences in the CLs calculated with different methods, waterbodies where it was possible to 5 6 use multiple methods were compared. There are three main CL approaches all based on 7 watershed mass-balance approach where acid-base inputs are balanced. The three approaches 8 include: (1) SSWC model and F-Factor that is based on quantitative relationships to water 9 chemistry (Dupont et al. 2005, Scheffe et al. 2014, Lynch et al. 2022), (2) Statistical Regression Model that extrapolated weathering rates across the landscape using water quality or landscape 10 11 factors (Sullivan et al. 2012a and McDonnell et al. 2014), and (3) Dynamic Models (MAGIC or Pnet-BGC). Critical load values were compared between these models to determine model 12 biases. Results from the comparison between different CL methods that were used to calculate 13 14 the critical loads in the NCLD are summarized in Appendix 5A, section 5A.3.1 for lakes in New 15 England and the Adirondacks and streams in the Appalachian Mountains. Overall, good agreement was found between the three methods used to calculate CLs, indicating there was not 16 a systematic bias between the methods and that they should produce comparable results when 17 18 used together as they were in these analyses.

#### 19 5.2.5 Summary

20 Quantitative analyses were performed to assess acidification risks of S deposition in 21 waterbodies across the U.S. using a critical load approach. Due to the finding of a negligible 22 influence of N deposition on acidification under current deposition levels, we focused on S 23 deposition solely. For these analyses ANC was used as the water quality indicator of 24 acidification, based on its longstanding use for this purpose. We also focused on acid-deposition sensitive areas for which the available CL modeling estimates indicated that the target ANC 25 26 values of 50, 30 and 20 µg/L could be reached. Analyses were performed at three different 27 spatial scales: nationwide, ecoregion III, and case studies. The results of these analyses are 28 summarized with regard to spatial extent and severity of deposition-related acidification effects 29 and the protection from these effects associated with a range of annual S deposition. Between the three-year period 2000-2002, which was the analysis year for the 2011 REA, 30 and 2018-2020, the latest period considered in the present analyses, national average sulfur 31 deposition has declined by 68% across the U.S. This decline in deposition is reflected in the very 32 different aquatic acidification impact estimates for the two periods. Unlike the findings for 2000-33 2002 in the last review (concluded in 2012), few waterbody sites are estimated to be receiving 34

35 deposition in excess of their critical loads for relevant ANC targets under recent deposition

1 levels. While recognizing inherent limitations and associated uncertainties of any such analysis,

2 the national scale assessment performed as part of this review, indicates that under deposition

3 scenarios for the 2018-2020 time period, only about 4% of waterbodies nationwide would not be

- 4 able to maintain an ANC of 50  $\mu$ g/L in the east and an ANC of 20  $\mu$ g/L in the west (see Table 5-
- 5 1).

6 The ecoregion-level analyses of ANC levels and deposition estimates for the five periods 7 from 2001-2003 through 2018 -2020 illustrate the spatial variability and magnitude of the impacts that might be expected for several target ANC levels (50, 30 and 20  $\mu$ g/L), and the 8 9 temporal changes across the 20-year period. For example, during the two most recent 3-year periods, the ecoregion median S deposition estimates in 2014-16 were below 5 kg/h-yr in all 10 11 ecoregions and the estimates for 2018-20 were all below 4 kg/h-yr. In this analysis, we summarized the ecoregion-level exceedances of CLs for each of the ANC targets in each of the 12 five time periods. While recognizing limitations and associated uncertainties of these analyses, 13

14 we note several key observations.

15 Although the ecoregion S deposition estimates in the 18 eastern ecoregions analyzed 16 were all below 5 kg/ha-yr in the two most recent time periods (2014-16 and 2018-20), the full dataset of five time periods a range from below 2 up to nearly 18 kg/ha-yr. Across this dataset of 17 18 CL exceedances for the three ANC targets for all 90 eastern ecoregion-time period combinations, 19 73% of the combinations had at least 90% of waterbodies per ecoregion estimated to achieve 20 ANC at or above 20 µeq/L, and 60% had at least 90% of the waterbodies estimated to achieve ANC at or above 50 µeq/L. The much higher deposition levels of the past are evident by the fact 21 22 that fewer than half of the eastern ecoregion-time period combinations (and all of the western 23 combinations) had an S deposition estimate below 4 kg/ha-yr.

24 Ninety percent of the eastern ecoregion-time period combinations were for ecoregion 25 deposition estimates at or below 13 kg/ha-yr. For these combinations (at or below 13 kg/ha-yr), 26 at least 90% of waterbodies per ecoregion were estimated to achieve an ANC at or above 20, 30 and 50 µeq/L in 80, 73 and 65% of all ecoregion-time period combinations, respectively. For S 27 deposition estimates at or below 9 kg/h-yr (approximately three quarters of the combinations), at 28 29 least 90% of all waterbodies per ecoregion were estimated to achieve ANC at or above 20, 30 and 50 µeq/L in 87, 81 and 72% of combinations. respectively. For S deposition estimates at or 30 below 5 kg S/h-yr, these values are 96, 92 and 82% of combinations. For the 75 western 31 ecoregion-time period combinations, all of which had an S deposition estimate below 4 kg/ha-yr, 32 33 at least 90% of waterbodies per ecoregion were estimated to achieve an ANC at or above 50 34  $\mu g/L.$ The case study analyses provide estimates of S deposition that might be expected to allow 35

36 these geographically diverse locations, including several Class I areas, to meet the three ANC

1 targets. In reviewing these estimates, we recognize inherent limitations and associated

- 2 uncertainties. Focusing on the three eastern case studies, the CL modeling indicates that at an
- annual average S deposition of 9-10 kg/h-yr, the sites in these areas, on average, might be
- 4 expected to achieve an ANC at or above 50 µeq/L. At an annual average S deposition of about 6-
- 5 9 kg/h-yr, 70% of the sites in the areas are estimated to achieve an ANC at or above 20  $\mu$ eq/L
- 6 and at about 5-8 kg/h-yr, 70% are estimated to achieve an ANC at or above 30  $\mu$ eq/L. Lower S
- 7 deposition values are estimated to achieve higher ANC across more sites. Across the three
- 8 eastern areas, the CL estimates for each ANC target are lowest for the White Mountains National
- 9 Forest study area, and highest for the Shenandoah Valley study area.

# 10 5.3 NITROGEN ENRICHMENT

There are several other categories of effects to aquatic ecosystems from deposition of 11 12 nitrogen and sulfur for which there is significant scientific evidence and causality judgements, as described in Chapter 4. These include N enrichment in various types of aquatic systems, 13 including freshwater streams and lakes, estuarine and near-coastal systems, and wetlands, as 14 described in section 4.2.2.1 above.<sup>3</sup> Separate quantitative analyses were not performed for these 15 categories of effects in this review. As recognized above, quantitative analyses have been 16 performed for welfare endpoints for which the evidence is most robust, and for which the 17 available information, tools and assessment approaches is supportive of such analyses for the 18 purposes in this review. With regard to the effects related to N enrichment in various types of 19 aquatic ecosystems, such analyses were not performed due to recognition of a number of factors, 20 21 including modeling and assessment complexities, and site- or waterbody-specific data 22 requirements, as well as, in some cases, issues of apportionment of atmospheric sources separate from other influential sources. Quantitative information relating deposition to consideration of 23 ecosystem effects has been described below for two of these categories, for which the ISA 24 25 summarizes studies that have developed critical load estimates. These categories are effects 26 related to N enrichment in wetlands and freshwater lakes and streams.

#### 27 5.3.1 Wetlands

Significant new information has become available since the 2008 ISA on N critical loads
for U.S. wetlands. While critical loads have previously been identified for European wetlands
such as bogs, fens, and intertidal wetlands for a variety of endpoints (growth, species

31 composition, species competition, peat and peat water chemistry, decomposition, and nutrient

<sup>&</sup>lt;sup>3</sup> Two other categories of effects assessed in the ISA (and for which causal determinations are made) are mercury methylation, and sulfide toxicity (ISA, Appendix 12). These categories of effects are described in section 4.2.3 above,

1 cycling) (Bobbink et al., 2003), recent studies have shown that CLs for sphagnum moss effects in

- 2 European bogs may not be directly relevant or transferrable to North American and/or U.S.
- 3 wetlands (ISA, Section 11.3). In U.S. coastal wetlands, two studies are available that have

4 considered N loads below 100 kg N/ha/yr. Wigand et al. (2003) estimated a critical load to

5 protect the community structure of salt marshes to be 63 to 400 kg N/ha/yr. Caffrey et al. (2007)

6 provided additional evidence that 80 kg N/ha/yr can alter microbial activity and

7 biogeochemistry. Two recent studies have described CLs for effects in freshwater wetlands. A

8 CL for wetland C cycling, quantified as altered peat accumulation and net primary productivity,

9 has been estimated between 2.7 and 13 kg N/ha/yr (Greaver et al 2011). A critical load for purple

10 pitcher plants (*Sarracenia purpurea*) has also been estimated (between 6.8-14 kg N/ha/yr) to

11 protect the population based on morphology and population dynamic endpoints.

12 A comparison of freshwater wetland CLs to observed ecological impacts of N from

recent studies (4.4–500 kg N/ha/yr) is provided in the ISA (Appendix 11, Figure 11-7). At the

14 lowest experimental addition level (16 kg N/ha/yr), there are observations of altered C and N

15 cycling and altered biodiversity. The endpoints affected include decreases in moss cover,

16 increased peat biomass, decreased N retention efficiency, and altered/damaged leaf stoichiometry

in vascular plants. However, this information is limited, and additional experimental evidence is

18 needed on critical loads for North American wetlands.

19 5.

## 5.3.2 Freshwater Lakes and Streams

20 Since the 2008 ISA, empirical and modeled critical loads for the U.S. have been 21 estimated based on surface water  $NO_3$  concentration, diatom community shifts, and 22 phytoplankton biomass growth nutrient limitation shifts. A critical load ranging from 3.5 to 6.0 kg N/ha/yr was identified for high-elevation lakes in the eastern U.S. based on the nutrient 23 24 enrichment inflection point (where NO3<sup>-</sup> concentrations increase in response to increasing N 25 deposition). Another critical load of 8.0 kg N/ha/yr was estimated by Pardo et al. (2011) for eastern lakes based on the value of N deposition at which significant increases in surface water 26 27 NO<sub>3</sub>- concentrations occur. In both Grand Teton and Yellowstone national parks, critical loads for total N deposition ranged from  $<1.5 \pm 1.0$  kg N/ha/yr to  $>4.0 \pm 1.0$  kg N/ha/yr (Nanus et al., 28 29 2017). Exceedance estimates were as high as 48% of the Greater Yellowstone area study region, 30 depending on the threshold value of NO<sub>3</sub>- concentration in lake water selected as indicative of biological harm. 31

Additional critical loads have been identified since the 2008 ISA for eastern Sierra
 Nevada lakes, Rocky Mountain lakes, the Greater Yellowstone Ecosystem, and Hoh Lake,

34 Olympic National Park (ISA, Appendix 9, Table 9-4). The identified values fall near or within

the range of 1.0 to 3.0 kg N/ha/yr for western lakes (Baron et al., 2011). An empirical critical

1 load of 4.1 kg/TN/ha/yr above which phytoplankton biomass P limitation is more likely than N

2 limitation was identified by Williams et al. (2017) for the western U.S. Modeled critical loads

3 ranged from 2.8 to 5.2 kg/TN/ha/yr, and a performance analysis indicated that a critical load of

4 2.0 kg/TN/ha/yr would likely reduce the occurrence of false negatives to near zero. However,

5 this evidence is geographically specific perhaps even waterbody specific and is not available for

- 6 most of the U.S.
- 7

### 5.4 TERRESTRIAL ECOSYSTEMS

8 As noted in the introduction to this chapter, quantitative analyses in the 2012 N oxides/ 9 SO<sub>X</sub> review that related atmospheric deposition in recent times (e.g., since 2000) to terrestrial effects, or indicators of terrestrial ecosystem risk, were generally considered to be more 10 uncertain than conceptually similar modeling analyses for aquatic ecosystems (e.g., "aquatic 11 12 acidification is clearly the targeted effect area with the highest level of confidence" [2009 REA, section 7.5]; 2011 PA, section 1.3). The terrestrial quantitative analyses in that review were 13 comprised of a critical load-based quantitative modeling analysis focused on BC:Al ratio in soil 14 15 (the benchmarks for which are based on laboratory responses rather than field measurements) and a qualitative characterization of nutrient enrichment (2009 REA). A more qualitative 16 approach was taken for nutrient enrichment in the 2012 review by simply describing deposition 17 ranges identified from observational or modeling research as associated with potential 18 effects/changes in species, communities and ecosystems and recognizing the uncertainties 19 20 associated with quantitative analysis of these depositional effects (2011 PA, section 3.2.3). 21 In this review, rather than performing new quantitative analyses focused on terrestrial 22 ecosystems, we have taken the approach of drawing on prior analyses and published studies recognized in the ISA that provide information pertaining to deposition levels associated with 23 effects related to terrestrial acidification and N enrichment. We reached this decision in 24 consideration of the available studies and with investigation into various assessment approaches. 25 26 As described in section 5.2 above, a full quantitative assessment has been performed, at multiple 27 scales, for consideration of aquatic acidification, an endpoint for which the available information, tools and assessment approaches provides strong support of such analyses that are 28 targeted to the needs in this review. For terrestrial effects related to N and S deposition, this 29

section draws on quantitative information relating deposition to consideration of terrestrial
 ecosystem effects, as described below and in the following subsections.

Since the 2012 N oxides/ SO<sub>X</sub> review, in addition to publications that apply steady-state (and dynamic) modeling to predict future soil acidity conditions in various regions of the U.S. under differing atmospheric loading scenarios, several publications have analyzed large datasets from field assessments of tree growth and survival, as well as understory plant community richness, with estimates of atmospheric N and/or S deposition. These studies investigate the
existence of associations of variations in plant community or individual measures (e.g., species
richness, growth, survival) with a metric for deposition during an overlapping time period,
generally of a decade or two in duration. Both mass balance modeling and observational studies,
as well as experimental addition studies, are, to various extents, informative in considering N and
S deposition levels of interest in the review.

7 In general, observational or gradient studies differ from the chemical mass balance 8 modeling approach in a number of ways that are relevant to their consideration and utilization for 9 our purposes in this review. One difference of note is the extent to which their findings reflect or 10 take into account the ecosystem impacts of historical deposition. Observational studies are 11 describing variation in indicators in the current context (with any ecosystem impacts, including stores of deposited chemicals that remain from historical loading). Historical loading, and its 12 associated impacts, can also contribute to effects analyzed with estimates of more recent 13 14 deposition in observational studies. Mass balance modeling, in the steady-state mode that is 15 commonly used for estimating critical loads for acidification targets, does not usually address the complication of historical deposition impacts that can play a significant role in timing of system 16 17 recovery.

18 For example, in considering the potential for terrestrial ecosystem impacts associated with different levels of deposition, the simple mass balance models common for estimating 19 20 critical acid loads related to BC:Al ratio are often run for the steady state case. Accordingly, the underlying assumption is that while historic deposition, and the various ways it may affect soil 21 22 chemistry into the future (e.g., through the stores of historically deposited sulfur), may affect 23 time to reach steady state (e.g., as the system processes the past loadings), it would not be 24 expected to affect the steady state solution (i.e., the estimated critical load for the specified soil 25 acidification indicator target). The complexities associated with site-specific aspects of 26 ecosystem recovery from historic depositional loading become evident through application of 27 dynamic models.

Observational studies, on the other hand, are inherently affected by historical deposition 28 29 and any past or remaining deposition-related impacts on soil chemistry and/or biota, in addition to other environmental factors. The extent of the influence of historical deposition (and its 30 ramifications) on the associations reported in these studies with metrics quantifying more recent 31 deposition is generally not known. Where patterns of spatial variation in recent deposition are 32 similar to those for historic deposition, it may be reasonable to conclude, however, that there is 33 34 potential for such influence. This is an uncertainty associated with interpretation of the observational studies regarding the deposition levels responsible for the observed variation in 35 plant or plant community measures. Thus, while observational studies contribute to the evidence 36

1 base on the potential for N/S deposition to contribute to ecosystem effects (and thus are

- 2 important evidence in the ISA determinations regarding causality), they may be somewhat less
- 3 informative with regard to identification of specific N and S deposition levels that may elicit

4 ecosystem impacts of interest. Both types of studies are considered in the sections below.

5 5.4.1 Soil Chemistry Response

6 Quantitative linkages between N and S deposition and soil chemistry responses vary 7 across the geography of the U.S. As summarized in sections 4.2.1.2 and 4.2.2.2 above, 8 acidification and N enrichment processes can alter the biogeochemistry in terrestrial ecosystems (ISA, Appendix 4). There are several indicators of acidification and N enrichment that also have 9 linkages to biological responses that are commonly used in quantitative analyses (ISA, Appendix 10 4, Table 4-1). These indicators are soil characteristics strongly associated with specific aspects of 11 soil acidification or nutrient enrichment. Uncertainties in the estimates of these indicators in 12 quantitative analyses for specific areas will generally be associated with limitations in the 13 estimation approach and the associated parameter values for those locations. 14

A commonly used indicator for soil acidification in quantitative modeling analyses of the 15 effect of acidifying deposition on forests (see section 5.3.2 below) is the ratio of base cations to 16 aluminum (BC:Al), with higher ratios indicating a lower potential for acidification-related 17 18 biological effects (ISA, Table IS-2). The ratio can be reduced by release of base cations from the 19 soil (e.g., through the process of neutralizing drainage water acidity) which reduces the base saturation of the soil. Soil base saturation<sup>4</sup> and changes to it can also be an indicator of 20 acidification risk (ISA, Appendix 4, section 4.3.4). The accelerated loss of base cations through 21 22 leaching, decrease in base saturation, and decreases in soil solution Ca:Al ratio all serve as 23 indicators of soil acidification. Inorganic and organic acids can be neutralized by soil weathering 24 or base cation exchange, in addition to denitrification (ISA, Appendix 4, section 4.3).

There are many indicators of N enrichment and potential eutrophication, including N accumulation, e.g., increased soil N concentrations or decreased C:N ratios (ISA, section IS.5.1.1). Increases in soil N can, however, also lead to nitrate leaching, potentially imposing a drain on base cations and a potential for increased acidity (ISA, Appendix 4, section 4.3). Thus, nitrate leaching can be an indicator of potential for increased aquatic acidity, as well as for terrestrial (or aquatic) N enrichment. Studies in various locations throughout the eastern U.S. and in the Rocky Mountains have reported estimates of N deposition associated with an onset of

32 increased nitrate leaching (ISA, Appendix 4, sections 4.3.2 and 4.6.2). For example, based on

<sup>&</sup>lt;sup>4</sup> As described in the ISA, "[s]oil base saturation expresses the concentration of exchangeable bases (Ca, Mg, potassium [K], sodium [Na]) as a percentage of the total cation exchange capacity (which includes exchangeable H+ and inorganic Al)" (ISA, Appendix 4, p. 4-27).

1 monitoring results for an 8-year experimental addition experiment in an alpine dry meadow in

2 the Rocky Mountains, with annual additions of 20, 40 and 60 kg N/ha-yr (Bowman et al., 2006),

3 Bowman et al., (2014) reported 10 kg N/ha/yr to be associated with enhanced nitrate leaching at

4 this location (ISA, Appendix 4, section 4.6.2.2).

Thus, the response of a terrestrial system, and the associated biota, to N additions as 5 6 through atmospheric deposition, can be one of acidification or nutrient enrichment depending on 7 the geology and soil chemistry (e.g., base cation weathering rate or base cation exchange capacity), residual impacts of historic deposition (e.g.,  $SO_4^{2-}/NO_3$  stored in soil) and organic 8 content, as well as acid sensitivity or growth limitations of the resident species. With regard to 9 soil indicators of nutrient enrichment (i.e., levels associated with particular risk of harm or 10 degree of protection), there is little research in the U.S. on which to base target values for 11 indicators such as soil N accumulation or NO<sub>3</sub> leaching (Duarte et al., 2013). This and 12 uncertainties associated with site-specific characteristics (e.g., carbon and organic content of 13 14 soils) may affect the use of soil modeling for identifying deposition targets aimed at controlling 15 nutrient enrichment.

#### 16 5.4.2 Effects on Trees

In this section we summarize the findings related to quantitative evaluation of S and N 17 18 deposition effects on trees. While S deposition contributes to acidification and its associated 19 negative effects on terrestrial systems, N deposition, as described in Chapter 4 and section 5.4.1 20 above, may contribute to acidification and/or nutrient enrichment, with associated effects on tree 21 growth and survival that, for acidification, can be negative, and, for nutrient enrichment, can be 22 positive or negative (ISA, Appendix). While the response is influenced by site-specific 23 characteristics, some species-specific patterns have also been observed (ISA, Appendix 6, 24 section 6.2.3.1). For example, conifer species, particularly at high elevations, were more likely to exhibit negative growth responses or mortality in response to added N and less likely to 25 26 demonstrate increased growth (ISA, Appendix 6, section 6.2.3.1; McNulty et al., 2005; Beier et 27 al., 1998; Boxman et al., 1998a). Variation in response can also be related to site-specific factors 28 contributing to variations associated with location. For example, while some long-term N 29 addition experiments indicate that broadleaf species more commonly exhibit increased growth (than conifers), there is variation across studies as seen in Appendix 5B (Table 5B-1). The extent 30 to which species-specific observations are related to the site-specific characteristics of areas 31 where species are distributed or to species-specific sensitivities is not clear. 32 33 In the subsections below, we draw on three main categories of studies: steady-state mass 34 balance modeling, experimental addition studies and observational or gradient studies. As noted

in section 5.4. above, each of these categories of studies has associated strengths and

limitations/uncertainties for our purposes here. For example, while the mass balance modeling
 studies are explicitly focused on acidic deposition effects, observational studies, given their real world settings, may reflect patterns of deposition contributing to both acidic deposition and/or
 the effects of nutrient enrichment. Thus, the subsections below are organized by study category

5 within which the findings with regard to both types of effects are discussed.

#### 6 5.4.2.1 Steady-State Mass Balance Modeling

As for assessment of aquatic acidification (see section 5.2 above), steady-state mass
balance modeling is also utilized to identify N/S deposition rates associated with conditions
posing differing risks to tree health. The evidence base evaluating such modeling, however, is
somewhat less robust than for aquatic ecosystems, such that the foundation for identifying target
conditions for neutralizing acidification, and for identifying appropriate values for some model
parameters, is more limited and uncertain, as noted below.

The indicator most commonly utilized to identify conditions associated with protection 13 from acidifying deposition risks to tree growth and survival is BC:Al (ISA, Appendix 5, section 14 5.2.1). Two meta-analyses are often referenced to inform interpretation of estimated BC:Al ratios 15 16 with regard to associated potential risks to tree health: Sverdrup and Warfvinge (1993) and Cronan and Grigal (1995). The first analysis compiled findings from laboratory, greenhouse and 17 field studies, with growth matrices varying from water solution to sand to field soil (Sverdrup 18 and Warfvinge (1993).<sup>5</sup> The literature review by Cronan and Grigal (1995), which reported the 19 20 Ca:Al ratios in 35 studies in which a response in seedling roots (e.g., change in nutrient content) were reported, is also often cited as a basis for selection of a target BC:Al value for use in simple 21 mass balance models. Nearly all of the 35 studies were conducted in hydroponic or sand systems, 22 in which aluminum is generally more freely available than in a soil substrate (Cronan and Grigal, 23 1995). As would be expected, there are limitations and uncertainties associated with findings 24 involving artificial substrates and growing conditions (ISA, Appendix 5, section 5.2.1).<sup>6</sup> In 25

consideration of these analyses, the BC:Al targets used in the 2009 REA for identifying

<sup>&</sup>lt;sup>5</sup> Ratios of BC:Al were identified using the cumulative percentage of experiments for tree seedling species grown in solution reporting a 20% growth reduction (Sverdrup and Warfvinge, 1993). For example, at cumulative percentage of 50% the BC:Al ratio was 1.2, and at 100% the ratio was on the order of 8 (Sverdrup and Warfvinger, 1993). The 2009 REA concluded that this analysis reported critical BC:Al ratios ranging from 0.2 to 0.8 (2009 REA, p. 4-54).

<sup>&</sup>lt;sup>6</sup> Based on the distribution Ca/Al ratios in the studies, Cronan and Grigal (1995) estimated a 50% risk of tree growth response for a molar ratio of 1.0 based on fact that 17 of the 35 studies had ratio at/above 1.0. The percentage of studies with a ratio at/above 1.8 was 25%., and it was approximately 5% at a ratio of 5, based on there being 33 of 35 or 94% of studies reporting a response for a Ca/Al ratio above 5. Only two of the 35 studies, both in conifers, reported a response, a change to root nutrient content (Cronan and Grigal (1995). In this assessment, "plant toxicity or nutrient antagonism was reported to occur at Ca/Al ratios ranging from 0.2 to 2.5" (2009 REA, p. 4-54).

- 1 acidifying deposition loads that might provide different levels of protection range from less than
- 2 1 to 10. Use of such target values (of 0.6, 1 and 10) in steady state simple mass balance modeling
- 3 in the last review resulted in the identification of acidifying deposition loads ranging from 487 to
- 4 2009 eq/ha-yr, across two areas of the Northeast for BC:Al target values differing by a factor of
- 5 nearly 20 (Table 5-6 and Table 5-7).

# Table 5-6. Acid deposition levels estimated for BC:Al targets in 24-state range of red spruce and sugar maple using steady-state simple mass balance model (2009 REA).

Target	Critical Loads for Acid Deposition for Different BC:Al Targets						
BC:AI	In terms of S+N (eq/ha-yr)	In terms of S (Kg S/ha-yr)	In terms of N (Kg N/ha-yr)				
0.6	1237-2009	40-64	17-28				
1	892-1481	29 - 48	13-21				
10	487-910	16- 29	7-13				

The 2009 REA (that informed the 2012 review of the NAAQS for N oxides and SO<sub>X</sub> 9 review) used the Simple Mass Balance (SMB) model for forest soil acidification, in steady-state 10 mode, to assess the extent to which atmospheric S and N deposition for the year 2002 might be 11 expected to contribute to soil acidification of potential concern (with BC:Al ratio used as an 12 13 indicator) for the sensitive species of sugar maple and red spruce in areas of 24-states where they are native (2011 PA, section 3.1.3; 2009 REA, section 4.3). The critical load analysis for the 14 three target BC:Al ratio values (identified for different levels of risk for growth impacts) drawn 15 from an estimated relationship between tree growth effect for different species and BC:Al ratio 16 17 yielded an array of estimates of acidifying deposition with potential to affect the health of at least 18 a portion of the sugar maple and red spruce growing in the United States (2009 REA, section 4.3 and Appendix 5; 2011 PA). 19 20 In addition to the uncertainty associated with characterization of risk for target BC:Al 21 ratio values, uncertainties were recognized in the SMB model calculations for the 2009 REA

analyses. For example, uncertainty recognized with the findings related to the use of default
 values for several key parameters (e.g., denitrification, nitrogen immobilization, the gibbsite

- equilibrium constant and rooting zone soil depth), and dependence of the SMB calculations on
- assumptions made in its application (2009 REA, section 4.3.9). Similarly, the ISA discussion of
- 26 SMB equations summarized findings of Li and McNulty (2007), who found uncertainty to come
- 27 primarily from components of the estimates for base cation weathering and acid-neutralizing
- capacity (ISA, section 4.5.1.2).

- Since the 2009 REA, an updated approach to estimating one particularly influential 1 parameter in the soil BC:Al modeling (cation weathering) has been reported (Phelan et al., 2 3 2014). Use of the new approach at 51 forested sites in Pennsylvania yielded rates consistent with soil properties and regional geology. The updated rates were generally higher, indicating a 4 greater buffering capacity for sites in this area to acidifying deposition than previously 5 determined (Phelan et al., 2014). The recent study by Duarte et al. (2013) also used updated 6 7 values for cation weathering for a study extending across New England and New York. For a soil BC:Al target of 10, this study reported a range of deposition estimates slightly higher than those 8
- 9 from the 2009 REA (see Table 5-7 below).

10Table 5-7.Acidic deposition levels estimated for several BC:Al ratio targets by steady-11state mass balance modeling for sites in northeastern U.S.

Endpoint, Species, Location	Deposition/Addition (loading)	Notes	
Modeling Analyses - Steady-state mass balance			
Range of risk for reduced growth (sugar maple and	487 to 2009 eq/ha-yr (7-28 kg N/ha-yr or 16-64	2009	
red spruce) in areas of 24 states in Northeast,	kg S/ha-yr)	REA	
based on soil BC:Al targets of 0.6, 1 and 10			
Soil BC:AL target of 10 for forest protection at	For a BC:Al target of 10, 850-2050 eq/ha-yr (27-	Duarte	
>4000 plots in New England and New York.	66 Kg S/ha-yr or 12-29 kg N/ha-yr), range for	et al.	
	80% of sites (for a BC:Al target of 10) total range	(2013)	
	was 11 to 6,540 eq ha-1yr-1, the lowest loads in		
	Maine, NH and VT		

# 12 5.4.2.2 Experimental Addition Studies

A number of experimental addition studies, conducted primarily in the eastern U.S., have 13 reported mixed results for growth and survival (see Appendix 5B, Table 5B-1). The species 14 studied have included oaks, spruce, maples, and pines. (Magill et al., 2004; McNulty et al., 2005; 15 Pregitzer et al., 2008; Wallace et al., 2007). Some multiyear S or N addition experiments 16 (involving additions greater than 20 kg/ha-yr) with a small set of eastern species, including sugar 17 maple, aspen, white spruce, yellow poplar, black cherry, have not reported tree growth effects 18 19 (ISA, section 5.5.1; Bethers et al., 2009; Moore and Houle, 2013; Jung and Chang, 2012; Jensen et al., 2014). Studies described in Appendix 5B are summarized here, including the annual 20 21 amounts of N added (in addition to the background deposition occurring during these times): 22 • Additions of 25 to as high as 150 kg N/ha-yr for 8-14 yrs (dating back to 1988) was associated with increased growth reported in sugar maple and oaks, at sites in MI, MA, 23 24 NY, ME.

Additions of 15.7 and 31.4 kg N/ha-yr for 14 yrs (beginning 1988) was associated with
 reduced basal area (red spruce) or growth (red maple, tulip poplar and black cherry, red
 pine) at sites in VT, MA, WV.

- Additions of 25 kg N/ha-yr for 13 yrs (beginning in 1989) was associated with increased growth rates for sugar maple but not for red spruce.
- 3 The N deposition levels simulated in experimental addition studies that report tree effects,
- 4 (including either increased or reduced growth, are generally greater than 10 kg N/ha-yr
- 5 (Appendix 5B, Table 5B-1).

### 6 5.4.2.3 Observational or Gradient Studies

Since the last review of the NAAQS for N oxides and SO<sub>X</sub>, several observational studies 7 have been published that investigate the existence of statistical associations between tree growth 8 or survival, as assessed at U.S. Forest Service, Forest Inventory and Analysis program 9 (USFS/FIA)<sup>7</sup> sites across the U.S., and estimates of average deposition of S or N compounds at, 10 averaged over multiyear time periods (Appendix 5B, section 5B.2.2; ISA, Appendix 5, section 11 5.5.2 and Appendix 6, section.6.2.3.1; Dietz and Moorcroft, 2011; Horn et al., 2018). The 12 standardized protocols employed in the FIA program make the use of the FIA plot data a strength 13 14 of these studies. These studies generally utilized the tree measurement data collected by the USFS from periodic assessments at each site, and data for other factors analyzed, including 15 metrics for atmospheric deposition (Table 5-8; Dietz and Moorcroft, 2011; Thomas et al., 2010; 16 Horn et al., 2018). 17 The study by Dietze and Moorcroft (2011) statistically evaluated the influence of a 18 number of factors, in addition to SO<sub>4</sub><sup>2</sup>-and NO<sub>3</sub> wet deposition (site-specific estimates of average 19 of 1994-2005 annual averages), on tree mortality (assessed over 5-15-year measurement intervals 20 21 within the period from 1970s through early 2000s) in groups of species characterized by

- functional type (267 species categorized into 10 groups) at sites in the eastern and central U.S.
- 23 (Appendix 5B, section 5B.2.2.1; ISA, Appendix 5, section 5.5.2). The full range of average  $SO_4^{2-}$
- 24 deposition was 4 to 30 kg S/ha-yr (Dietze and Moorcroft, 2011). Other factors assessed (which
- 25 were all found to have statistically significant associations with more than one of the tree species
- 26 groups) were precipitation, minimum and maximum temperature, ozone, topographic factors
- 27 (elevation, slope and variation in solar radiation and soil moisture), and biotic interaction factors
- 28 (stand basal area and age, and focal-tree diameter at breast height). The authors reported that the
- 29 strongest effect was due to acidifying deposition (specifically  $SO_4^{2-}$ ), particularly in the northeast
- 30 sites (Dietze and Moorcroft, 2011). Negative associations were reported with tree survival for 9
- of the 10 functional groups. Survival for the same 9 groups were also negatively associated with

<sup>&</sup>lt;sup>7</sup> The FIA Program's forest monitoring component involves periodic assessments of an established set of plots distributed across the U.S. This component includes collection of data at field sites (one for every 6,000 acres of forest). The data include forest type, site attributes, tree species, tree size, and overall tree condition. At a subset of the plots, a broader suite of forest health attributes including tree crown conditions, lichen community composition, understory vegetation, down woody debris, and soil attributes are also assessed (USFS, 2005).

1 long-term average ozone concentrations. The third highest influence was for N deposition (range

- 2 across sites was 6 to 16 kg N/ha-yr), with mortality in all but one species group having a
- 3 negative association (i.e., lower probability of mortality with higher NO<sub>3</sub> deposition). Regarding
- 4 the significant associations with S and N deposition, the authors recognized that "[t]he impacts
- 5 of both acidification and nitrogen deposition on tree mortality result from cumulative, long-term
- 6 deposition, and the patterns presented [in their paper] should be interpreted in that light" (Dietze
- 7 and Moorcroft, 2011).

8 The study by Thomas et al. (2010) focused on relationships of tree growth and survival 9 (assessed at FIA plots from 1978 through 2001, with measurement interval ranging from 8.3 to 14.4 years) with N deposition (mean annual average for 2000-04) as the only pollutant included 10 in the statistical analyses (Appendix 5B, section 5B.2.2.2). Increased growth was associated with 11 higher N deposition in 11 of 23 species in northeastern and north-central U.S and with lower N 12 deposition in 3 species (Thomas et al., 2010). Eight species showed negative associations of 13 14 survival rates with N deposition and three showed positive associations. The other factors 15 analyzed included temperature, precipitation, and tree size, but did not include other pollutants (Thomas et al., 2010). 16

The third study utilizing measurements at USFS plots, reported on statistical modeling of 17 tree growth and survival of 71 species at USFS plots across the U.S. with site-specific estimates 18 of average S and N deposition across the measurement interval (generally 10 years) within the 19 20 period from 2000-2013 (Horn et al., 2018; Appendix 5B, section 5B.2.2.3). The study focused on 21 71 of 94 species for which covariance between N and S deposition metric values and other 22 factors was a lower concern (Horn et al., 2018). Of the 71 species on which the analysis focused, negative associations were reported for survival and growth with S deposition estimates for 40 23 24 and 31 species, respectively. Sulfur deposition at sites of these species ranged from a minimum 25 below 5 kg/ha-yr to a site maximum above 40 kg/ha-yr, with medians for these species generally ranging from around 5 to 12 kg/ha-yr (Appendix 5B, section 5B.3.2.3). 26 27 The study by Horn et al (2018) also reported associations of growth and survival with N deposition estimates that varied positive to negative across the range of deposition at the 28 29 measurement plots for some species, and also among species (Horn et al., 2018). For the six

- 30 species, for which survival was negatively associated with the N deposition metric across the full
- range of values, the site-specific deposition metric ranged from below 5 to above 50 kg/ha-yr,
- 32 with medians ranging from 8 to 11 kg N/ha-yr (Appendix 5B, Figure 5B-7). The median values
- for the 19 other species with unimodal (or hump-shaped) associations that were negative at the
- 34 species median deposition value (and for which sites were not limited to the western U.S.)
- ranged from 7 to 11 kg N/ha-yr. The deposition metric ranges were generally similar for the
- 36 species for which survival was positively associated with metric (across full range or at the

1 median). Of the 39 species for which growth was significantly associated with N deposition, the

- 2 association was negative across the full range for two species (with sample sites predominantly
- 3 in the Atlantic coastal pine barrens and northern plains and forests, respectively). The median
- 4 deposition across sites for these two were nine and ten kg N/ha-yr (Appendix 5B, Figure 5B-5
- 5 and Attachment 2). The median deposition values for the two other species with hump shaped
- 6 functions that were negative at the median were seven and eight kg N/ha-yr, respectively
- 7 (Appendix 5B, Figure 5B-5).

Observational studies newly available in this review include two smaller studies in the 8 9 Adirondacks of New York that investigated relationships of forest plot characteristics with N and S deposition metrics. These locations are well documented to have received appreciable acidic 10 deposition over the past several decades. The studies report negative associations of forest health 11 metrics with N and/or S deposition metrics (see Appendix 5B, Table 5B-2). Also newly available 12 in this review are studies that analyzed potential for associations of tree growth of sensitive 13 14 species with temporal changes in SOx and/or NOx emissions. For example, a study by Soule 15 (2011) reported increased red spruce growth in North Carolina to be associated with reductions in emissions of SOx and N oxides from utilities in the southeastern U.S., among other factors, 16 over the period from 1974 to 2007 (Soule, 2011; ISA, Appendix 5, section 5.5.1). 17 Another observational study newly available in this review documented recovery of a 18 stand of eastern redcedar (in the Appalachian Mountains of West Virginia) from historical S 19

- pollution using an analysis of tree ring chronology from 1909 to 2008, and a multivariate
  correlation analysis involving historical climate variables, atmospheric CO<sub>2</sub> concentrations and
  U.S. emissions estimates for SO<sub>2</sub> and N oxides (ISA, section 5.2.1.3; Thomas et al., 2013). Tree
  growth has increased significantly since 1970 and the analysis indicates it is explained by
  increases in atmospheric CO<sub>2</sub> and NOx emissions and reductions in SO<sub>2</sub> emissions (ISA, section
- 5.2.1.3; Thomas et al., 2013). The authors described the response as an indirect result of
- reductions in acid deposition, while other researchers have suggested that, given the speed of the
- response, it may more likely be related to reduced gaseous SO<sub>2</sub> than acid deposition (ISA,
- 28 section 5.2.1.3).
- 29

# 1Table 5-8.Tree effects and associated S/N deposition levels from observational studies2using USFS/FIA data.

Endpoint, Species, Location	Deposition/Addition	Reference				
S Deposition Metric Analyses						
Survival in 7 of 10 species' groups in eastern and central U.S. negatively associated with SO <sub>4</sub> <sup>2</sup> deposition	SO <sub>4</sub> <sup>2</sup> -wet deposition estimates (average, 1994- 2005) varied 4 to 30 kg S/ha-yr across all sites.	Dietze and Moorcroft (2011)				
Survival in 40 species across U.S. was negatively associated with S deposition estimates.	Median average S deposition estimates (2000-16) for these species: 3 <sup>A</sup> to 12 kg S/ha-yr.					
Growth in 31 species across U.S. was negatively associated with S deposition estimates.	Median S deposition estimates for these species varied 4 <sup>A</sup> to 12 kg S/ha-yr, when western species are excluded.	Horn et al. (2018)				
N Dep	osition Metric Analyses					
Mortality in 1 species' group in eastern/central U.S. positively associated with NO <sub>3</sub> deposition Mortality in 9 of 10 species' groups in eastern and central U.S. negatively associated with NO <sub>3</sub> deposition (reduced mortality with increased NO <sub>3</sub> )	$NO_3$ wet deposition estimates (average, 1994-2005) varied from 6 to 16 kg N/ha-yr across all sites analyzed	Dietze and Moorcroft (2011)				
Survival of 8 species negatively associated with N deposition. Survival of 3 species positively associated with N deposition. Growth of 3 species (all conifers) negatively associated with N deposition, Growth of 11 of 24 species positively associated with N deposition,	Estimates of average N deposition across the full set of study sites ranged from 3 to 11 kg N/ha-yr for the period 2000-2004.	Thomas et al. (2010)				
Survival of 6 species was negatively associated with N deposition across deposition ranges Survival of 21 other species (2 limited to the West), with U-shaped associations, also negatively associated with N deposition at median deposition across species' sites. Survival of one species positively associated with N deposition across deposition range Survival of 4 other species, with U-shaped associations, also positively associated with N deposition at median deposition for species' sites. Growth of 2 species was negatively associated with N deposition across all species' sites. Growth of 2 other species (with U-shaped associations) also negatively associated with N deposition at the median deposition across sites Growth of 20 species (17 nonwestern species) was positively associated with N deposition across all species' sites. Growth of 15 other species with U-shaped associations (14 nonwestern species) was also	For species with negative associations, median N deposition estimates varied from 8 to 11 kg N/ha-yr. For 19 species with negative association at median deposition, western species excluded, median N deposition varied 7 to 12 kg N/ha-yr. For species with positive association, median N deposition estimate was 11 kg N/ha-yr. For species with positive association at median deposition, median N deposition varied from 7 to 12 kg N/ha-yr. The median average deposition estimates for the measurement interval (during 2000-16) varied from 9 and 10 kg N/ha-yr. The median estimates for the other 2 species were 7 and 8. The 17 nonwestern species assessed at sites for which the median average deposition estimate for the measurement interval (during 2000-16) varied from 7 to 12 kg N/ha-yr. The median estimates for the other 14 nonwestern species were 7 to 11 kg N/ha-yr	Horn et al. (2018)				
deposition across those species' sites.						

#### 1 5.4.3 Other Effects

The studies available that may inform consideration of S or N deposition levels of potential interest for deposition-related effects on terrestrial biota other than trees include both addition experiments and observational or gradient studies. In addition to effects on individual species, these studies often report metrics related to changes in communities of particular plant or lichen populations. Information from both types of studies and with regard to species-level or community-level effects are discussed in the subsections below. The focus in these studies, however, is predominantly on N deposition.

#### 9 5.4.3.1Effects on Herbs and Shrubs

#### 10 Observational/Gradient Studies

Since the 2012 review, new observational studies have investigated relationships between 11 deposition and community composition for understory plants. One of the largest studies, Simkin 12 et al. (2016), investigated relationships between species richness (number of species) of 13 herbaceous plants<sup>8</sup> and values of a N deposition metric at more than 15,000 forest, woodland, 14 shrubland and grassland sites across the U.S. (Appendix 5B, section 5B.4.2). The study grouped 15 the sites into open- or closed-canopy sites, with forest sites falling into the closed-canopy 16 category and the rest, open-canopy. The data for sites in each of the two categorized were 17 analyzed for relationships of species richness (number of herbaceous species) with values of the 18 N deposition metric, soil pH, temperature, and precipitation (Simkin et al., 2016). The species 19 richness assessments were conducted across a 23-year period (1990-2013) by multiple 20 researchers, at sites clustered most prominently in portions of the 14-state study area, e.g., MN, 21 WA, OR, VA, NC and SC (Appendix 5B, Figure 5B-13). The N deposition metric for each site 22 23 was a 10-year average of dry N deposition (2002-2011) added to a 27-year average (1985-2011) of wet deposition (Simkin et al., 2016; Appendix 5B, section 5B.4.2). 24 Different relationships among the analyzed factors were observed for the two categories 25 of sites, with a hump-shaped relationship of species richness with the deposition metric at open-26 27 canopy sites and a strong influence of soil pH at the closed-canopy (forest) sites (Simkin et al., 28 2016). • At open-canopy sites, the association of herbaceous species richness with the N deposition 29 metric was somewhat dependent on soil pH, precipitation and temperature. Herbaceous 30

metric was somewhat dependent on son pri, precipitation and temperature. Heroaccous
 species richness was positively associated with the N deposition metric at the lower end
 of the deposition range and negatively associated with N deposition at the higher end of
 the deposition range, on average for metric values above 8.7 kg N/ha/yr (Simkin et al.,
 2016).

<sup>&</sup>lt;sup>8</sup> Herbaceous plants are nonwoody vascular plants, including annuals, biannual and perennials.

- At closed-canopy (forest) sites, the association of herbaceous species richness with the N deposition metric was highly dependent on soil pH. Across sites with acid soil pH at/above 4.5, species richness was negatively associated with N deposition metric values greater than 11.6 kg N/ha/yr, but among sites with basic soils there was no point in the data set at which N deposition had a negative effect on species richness (the analysis included deposition values up to ~20 kg N/ha/yr).
- 7

7 The long time period over which the N deposition estimates are averaged in this study provides for an N deposition metric generally representative of long-term N deposition over a 8 9 time period of temporally changing rates, particularly in areas of the Midwest south to the Gulf, and eastward (e.g., ISA, Appendix 2, section 2.7). The impact of the differing time periods for 10 the wet versus dry deposition estimates, however, is unclear. Notably, the study did not consider 11 12 potential roles for other pollutants with a potential influence on the observations, including ozone and S deposition. Overall, the study by Simkin et al. (2016) indicates an effect of N deposition 13 on herbaceous species richness, with a number of uncertainties that limit interpretations 14 15 regarding identification of specific deposition levels of potential concern with regard to impacts

- 16 on herbaceous species number.
- 17 Studies in southern California, particularly in grassland or coastal sage scrub
- 18 communities, have investigated the role of past N deposition in documented alterations of
- 19 community composition and increases in the presence of invasive species (ISA, Appendix 6,
- section 6.3.6). In light of the changes in vegetation that have occurred in this area since the early
- 21 20<sup>th</sup> century, a recent study by Cox et al. (2014) utilized a landscape-level analysis in
- 22 investigating the risk of coastal sage scrub communities converting to exotic annual grasslands
- and potential associations with N deposition. These analyses further considered the factors that
- 24 might influence or facilitate community recovery. Results of these analyses indicated that
- 25 recovery of coastal sage shrub communities<sup>9</sup> from exotic grass invasion was most likely in sites
- with N deposition below 11.0 kg N/ ha/yr (in 2002, based on CMAQ modeling) and had
- 27 experienced relatively low invasion (Cox et al., 2014).

28 Experimental Addition Studies

29 Several addition studies have focused on California coastal sage scrub communities (ISA,

Appendix 6, section 6.3.6). A study of 13 years of 50 kg N/ha-yr additions reported no

- significant effects on plant cover for the first 11 years of the 13-year period (ISA, Appendix 6, p.
- 6-81; Appendix 5B, Table 5B-7). Community composition was changed after five years,
- reflecting changes in the relative abundance of dominant shrubs, and in the  $11^{\text{th}}$  through  $13^{\text{th}}$
- 34 years, increases in an exotic plant and decreases in one of the native shrubs were reported
- 35 (Vourlitis, 2017; Vourlitis and Pasquini, 2009).

<sup>&</sup>lt;sup>9</sup> Coastal sage scrub is a shrubland community that occurs in Mediterranean-climate areas in southern California.

- 1 Experimental addition experiments have also reported variable relationships between N
- 2 additions and impacts for herb or shrub communities (ISA, section 6.3; Appendix 5B, section
- 3 5B.4). For example, a study by Bowman et al. (2012) in a dry sedge meadow in Colorado
- 4 reported no shifts in species richness or diversity in response to N additions of 5, 10 and 30 kg/ha-yr,
- 5 but also found increases in cover of one species (*Carex rupestris*) that ranged from 34 to 125%
- 6 across the treatments (ISA, Appendix 6, section 6.3.4).
- 7 At Joshua Tree National Park in the Mojave desert of California, non-native grass biomass increased significantly at three of the four study sites receiving 30 kg N/ha/yr for two 8 9 years but experienced no significant change with an addition of 5 kg N/ha/yr (Allen et al., 2009). No significant change in community composition or species richness was reported in a semi-arid 10 grassland in Utah in response to smaller additions of 2, 5 and 8 kg N/ha-yr over two years (ISA, 11 Appendix 6, Table 6-21; McHugh et al., 2017). Much higher additions, of 10, 20, 34, 54 and 95 12 kg N/ha-yr over 23 years, in prairie grasslands resulted in reduced species richness. Ceasing 13 those additions after 10 years resulted in recovery of species number back to control numbers<sup>10</sup> 14
- 15 after 13 years (Clark and Tillman, 2008).

# 16 5.4.3.2 Effects on Lichen

- 17 The available information on N, S or PM exposure conditions associated with effects on
- 18 lichen is primarily focused on nitrogen species (available evidence summarized in the ISA,
- 19 Appendix 6, section 6.5.2). Limited information regarding effects of  $SO_x$  on lichen species is
- summarized in section 5.1.1 above, and the extent to which the effects relate to airborne  $SO_x$  (vs
- 21 associated acidic deposition) is not clear. Somewhat similarly, section 5.1.2 above summarizes
- the available information regarding N oxides exposure conditions, including associated
- 23 deposition, for which effects are reported on lichen species. We address below several
- 24 observational or gradient studies newly available in this review that analyzed relationships
- 25 between lichen community characteristics and N and/or S deposition metrics at sites in the
- 26 Northeast and Northwest (Table 5B-9; ISA, Appendix 5, section 5.5.1 and Appendix 6,
- 27 Appendix 6, section 6.5).
- In the northeastern U.S., past studies have concluded that in areas distant from industrial
  or urban sources, atmospheric deposition alters chemistry of tree bark (that provides substrate for
- 30 lichen species) through acidification or eutrophication (Cleavitt et al., 2011; van Herk, 2001;
- ISA, Appendix 6, section 6.2.3.3). A study of relationships between lichen metrics and metrics
- for annual and cumulative N and S deposition from 2000 to 2013 at plots in four Class I areas of
- the northeastern U.S. reported that "lichen metrics were generally better correlated with

<sup>&</sup>lt;sup>10</sup> Species number changes in control plots contributed to this finding (Clark and Tillman, 2008; Isbell et al., 2013).

cumulative deposition than annual deposition" (Cleavitt et al., 2015). Further, cumulative dry 1 2 deposition of S yielded the best fit to decreases in thallus condition, poorer community-based S Index values, and absence of many S-sensitive species, indicating a stronger role for legacy of 3 historical deposition than recent deposition patterns (Cleavitt et al., 2015). Across the years 4 studied, annual S and N deposition in the four areas declined, from roughly 6-15 kg S/ha-yr to 3-5 6 6 kg S/ha-yr and from roughly 4-15 kg N/ha-yr to 3-8 kg N/ha-yr (Cleavitt et al., 2015, Figure 4). 7 Two studies, newly available in the ISA, involve sites in the Northwest and focus on 8 assessing relationships between metrics for lichen community composition and estimated N 9 deposition. The study by Geiser et al. (2010) related lichen air scores assigned based on relative abundance of oligotrophic and eutrophic species in assessments (conducted from 1994 to 2002) 10 to N deposition metric values (based on 1990-99 average N deposition). The authors identified a 11 breakpoint between the third and fourth air scores which was associated with 33-43% fewer 12 oligotrophic species and 3 to 4-fold more eutrophic species than sites with scores in the "best" 13 14 bin; at sites reflecting this scoring breakpoint, total N deposition estimates ranged from 3 to 9 kg 15 N/ha-yr (Geiser et al., 2010). Using a different score or index to characterize lichen communities (based on assessments 1993-2011), Root et al. (2015) analyzed particulate N estimated from 16 speciated PM<sub>2.5</sub> monitoring data and throughfall N deposition estimated from lichen N content. 17 Several aspects of these studies complicate interpretation of exposure conditions, and 18 identification of N deposition levels associated with particular risks to lichen communities. For 19 20 example, the methods for utilizing N deposition differ from current commonly accepted 21 methods. There is also uncertainty regarding the potential role of other unaccounted-for 22 environmental factors (including ozone, SO<sub>2</sub>, S deposition and historical air quality and associated deposition). There is uncertainty concerning the independence of any effect of 23 24 deposition levels from residual effects of past N deposition. And there are few controlled N 25 addition experiments that might augment or inform interpretation of the findings of 26 observational/gradient studies (fumigation studies are summarized in section 5.1.2 above). Other 27 studies in Europe and Canada have not reported such associations with relatively large N

28 deposition gradients.

# 5.5 KEY FINDINGS AND ASSOCIATED UNCERTAINTIES AND LIMITATIONS

# 31 5.5.1 Aquatic Acidification

Key findings related to deposition levels associated with aquatic acidification, and more
 specifically to different waterbody buffering capacity targets, in terms of ANC, are summarized
 below.

5-64

1 • 2	The most widely used indicator of surface water acidification, and subsequent recovery under scenarios with lower acidifying deposition, is ANC.
3 • 4 5 6	Considerable new research on critical loads for acidification is available and both steady state and dynamic models have been used to generate ANC based critical loads for much of the U.S. Empirical studies have also identified a range of critical loads over a wide range of ANC levels for selected areas known to be sensitive to acidification.
7 • 8 9 10 11	Quantitative assessments were developed for this review to evaluate the impact of nitrogen and/or sulfur deposition on aquatic acidification across the U.S. using a CL approach. This relationship between acidifying deposition of nitrogen and sulfur; water chemistry changes reflected by changes in ANC; and waterbody health and biodiversity are the basis for the quantitative assessments.
12 • 13 14 15 16 17	Key design elements of the approach employed in the quantitative assessments include the spatial scales, water quality indicator of acidification, how to define the CL and exceedance parameters, data sources for deposition estimates, consideration of relative S and N contributions to acidifying deposition, consideration of ecosystem sensitivity and attainability of specific ANC targets and focus for quantitative uncertainty analyses. These elements of the analyses are summarized here:
18	– Spatial Scale: National, Ecoregion III, and Case Study (Class I areas)
19	– Chemical Indicator: ANC, with target values of 20, 30 and 50 $\mu$ eq/L
20	<ul> <li>Critical Load Sources: NCLD Database and empirical CL from ISA</li> </ul>
21 22 23	<ul> <li>Exceedance Calculation: CLs are exceeded where deposition is above the CL+ 3.125 meq S/m<sup>2</sup>-yr or 0.5 Kg S/ha/yr and are not exceeded where deposition is below the CL - 3.125 meq S/m<sup>2</sup>-yr or 0.5 Kg S/ha/yr.</li> </ul>
24 25	<ul> <li>Deposition Data Source and Time Periods: TDEP and three-year averages were calculated for these periods: 2001-03, 2006-08, 2010-2012, 2014-16 and 2018-20</li> </ul>
26 27	<ul> <li>Relative Contributions: Focus on S deposition CLs as analyses indicated negligible contribution to acidification from N under most conditions</li> </ul>
28 29	<ul> <li>Attainability of ANC targets: CLs&lt;0 and those areas for which deposition was not a driving factor were not used in the analyses</li> </ul>
30 • 31 32	Under recent (2018-2020) levels of S deposition, and available CL modeling, around 4% of waterbodies nationwide for which we have sufficient data are not expected to attain an ANC of 50 $\mu$ eq/L.
33 • 34 35	Ecoregion-level analyses of ANC-based CLs for the five periods from 2000-2002 through 2018-2020 provide further characterization of both spatial variability of acid sensitive waterbodies across the U.S. and the magnitude of deposition driven acidification impacts.
36 37 38	<ul> <li>In the western ecoregions, for which the ecoregion S deposition estimates were below 4 kg/h-yr, the analysis indicated an ANC at or above 50 µeq/L to be achieved in all five time periods.</li> </ul>
39 40 41	<ul> <li>Between the three-year period 2000-2002, which was the analysis year for the 2011 REA, and 2018-2020, the latest period considered in the present analyses, national average sulfur deposition has declined by 68% across the U.S. This</li> </ul>

1 2 3 4 5 6 7 8	decline in deposition is reflected in the very different aquatic acidification estimates for the two periods. Unlike the findings for 2000-2002 in the las (concluded in 2012), few waterbody sites are estimated to be receiving dep in excess of their critical loads for relevant ANC targets under recent depo levels. While recognizing inherent limitations and associated uncertainties such analysis, the national scale assessment performed as part of this revie indicates that under deposition scenarios for the 2018-2020 time period, al 96% of waterbodies nationwide would be able to maintain an ANC of 50 p	impact t review position sition of any w, pout ug/L
9	(see Table 5-1).	
10	<ul> <li>Although the ecoregion S deposition estimates in the 15 eastern ecoregion</li> </ul>	S
11	analyzed were all below 5 kg/ha-yr in the two most recent time periods (20	014-16
12	and 2018-20), ecoregion deposition estimates for the full dataset of five tir	ne
13	periods range from below 2 up to nearly 18 kg/ha-yr. Across this dataset o	fCL
14	exceedances for the three ANC targets for all 90 eastern ecoregion-time pe	eriod
15	ecoregion estimated to achieve ANC at or above 20 ueg/L and 60% had a	er t legst
17	90% of the waterbodies estimated to achieve ANC at or above 50 µeq/L.	The
18	much higher deposition levels of the past are evident by the fact that fewer	r than
19	half of the eastern ecoregion-time period combinations (and all of the west	tern
20	combinations) had an S deposition estimate below 4 kg/ha-yr. Ninety perc	ent of
21	the eastern ecoregion-time period combinations were for ecoregion deposi	tion
22	estimates at or below 13 kg/ha-yr. For these combinations (at or below 13	kg/ha-
23	yr), at least 90% of waterbodies per ecoregion were estimated to achieve a	n ANC
24	at or above 20, 30 and 50 $\mu$ eq/L in 80, 73 and 65% of all ecoregion-time p	eriod
25 26	three quarters of the combinations) at least 90% of all waterbodies per equ	aregion
20	were estimated to achieve ANC at or above 20, 30 and 50 µeg/L in 87, 81	and
28	72% of combinations. respectively. For S deposition at or below 5 kg S/h-	vr.
29	these values are 96, 92 and 82% of combinations.	
30 31 32 33 34 35	• The case study analyses of the CL modeling for waterbodies in those geographical diverse locations include several Class I areas. In the three eastern case studies, th modeling indicates that at an annual average S deposition of 9-10 kg/h-yr, the site these areas, on average, might be expected to achieve an ANC at or above 50 µeq an annual average S deposition of about 6-9 kg/h-yr, 70% of the sites in the areas estimated to achieve an ANC at or above 20 µeq/L. Lower S deposition values are	ly e CL s in /L. At are e
36	estimated to achieve higher ANC across more sites.	
37	There are three major areas that contribute uncertainties to the results: (1) the link	age
38	between the biological/ecosystem response and acidification, (2) the linkage between spe	cific
39	ecological impacts and the ecological indicator (ANC) and (3) the linkages between depo	sition
40	and ANC through the CL approach.	
41	The first, the linkage between acidifying deposition and the ecosystem response h	as been
42	well documented over 40+ years of evidence (ISA, Appendix 8). Associations have been	long
43	established between aquatic acidification (e.g. reduced pH, and elevated Al) and adverse	

1 ecosystem effects, including fish mortality, decreased species diversity, etc. (ISA, Appendix 8).

- 2 Variability in quantitative aspects of these associations, which generally relate to factors such as
- 3 climatological conditions, lake and stream size, other water quality parameters (e.g., dissolved
- 4 organic carbon, dissolved oxygen, etc), biological interactions, etc, complicate the quantitative
- 5 relationship of biological/ecological responses to acidification.
- 6 The second area of uncertainty is in associating specific levels of ANC with specific
- 7 biological/ecological effects. The water quality parameter, ANC, is the preferred indicator for
- 8 acidification because of its linear relationship with deposition driven acidification as opposed to
- 9 pH which is influenced by natural factors such as the level of dissolved  $CO_2$  in water. Surface
- 10 water levels of ANC, pH and Al are controlled by well-defined aquatic equilibrium
- 11 chemistry. While the relationships between ANC and ecological impacts is well-known, there is
- 12 uncertainty in our understanding of relationships between ANC and risk to native biota,
- 13 particularly in waterbodies in geologic regions prone to waterbody acidity. Such uncertainties
- relate to the varying influences of site-specific factors other than ANC.
- 15 The third point of uncertainty is associated with our understanding of the biogeochemical linkages between deposition and ANC, and determination of steady-state CLs. This by far is the 16 largest uncertainty and the one that is most difficult to characterize and assess. There is 17 18 uncertainty associated with parameters in the steady-state CL models. While the SSWS and other 19 CL models are well conceived and based on a substantial amount of research and applications 20 available in the peer reviewed literature, there is uncertainty associated with the availability of 21 the necessary data to support certain model components. The strength of the CL estimate and the 22 exceedance calculation relies on the ability of models to estimate the catchment-average base-23 cation supply (i.e., input of base cations from weathering of bedrock and soils and air), runoff, 24 and surface water chemistry. The uncertainty associated with runoff and surface water 25 measurements is broadly understood, however, the ability to accurately estimate the catchment supply of base cations to a water body is still difficult. This is important because the catchment 26 27 supply of base cations from the weathering of bedrock and soils is the factor with the greatest influence on the CL calculation and has the largest uncertainty (Li and McNulty, 2007). 28 29 Although the approach to estimate base-cation supply for the national case study (e.g., F-factor approach) has been widely published and analyzed in Canada and Europe, and has been applied 30 in the U.S. (e.g., Dupont et al., 2005 and others), the uncertainty in this estimate is unclear and 31 32 could be large in some cases. For this reason, an uncertainty analysis focused on this aspect of
- 33 state-steady CL modeling was performed (summarized in section 5.2.4 above).

#### 1 5.5.2 Other Aquatic Effects

2 Key findings related to deposition levels associated with other aquatic effects are 3 summarized below. There are several other effects to aquatic ecosystems from deposition of 4 nitrogen and/or sulfur for which there are a range of associated deposition levels. Most of these impacts are associated with nitrogen deposition but some, such as sulfide toxicity, are primarily 5 related to sulfur. The eutrophication of wetlands and other aquatic systems is primarily 6 7 associated with nitrogen inputs whether from deposition or other sources. The ranges of deposition associated with these effects is very broad and ranges from less than 1 kg N/ha/yr for 8 9 impacts to diatom communities in high elevation lakes to over 500 kg N/ha/yr in some N addition studies in wetlands. The information available on these types of impacts is sufficient for 10 causal determinations but often localized or otherwise limited for the purpose of quantitative 11 assessment relating deposition to waterbody response at an array of U.S. locations. For this 12 13 review, these impacts were considered from a qualitative perspective and contribute to the evidence base described in Chapter 4. 14

#### 15 **5.5.3 Terrestrial Effects**

Key findings related to ambient air concentrations and deposition levels associated with
 terrestrial effects discussed in prior sections are summarized below.

#### 18 5.5.3.1 Direct Effects on Plants and Lichens of Pollutants in Ambient Air

19 The evidence related to exposure conditions for direct effects of SOx, N oxides and PM in ambient air includes concentrations of SO<sub>2</sub> and NO<sub>2</sub> associated with effects on plants, 20 concentrations of NO<sub>2</sub> and HNO<sub>3</sub> associated with effects on plants and lichens and quite high 21 22 concentrations of PM that affect plant photosynthesis. With regard to SO<sub>2</sub>, while most studies are focused on visible foliar injury in sensitive plants (with exposures varying from 8 hours at 0.2 23 ppm SO<sub>2</sub> to repeated hourly concentrations of 0.4 ppm), laboratory studies have also reported 24 reduced photosynthesis for repeated exposures of 3 to 4.2-hours/day to concentrations on the 25 order of 0.25 to 0.5 ppm SO<sub>2</sub>, and reduced soybean yield after repeated multi-hour exposures to 26 27 0.19 ppm SO<sub>2</sub> (section 5.1.1 above). The evidence comes from an array of studies, primarily field studies for the higher concentrations associated with visible foliar injury and laboratory 28 studies for other effects. Uncertainties relate to the extent to which effects observed in controlled 29 30 laboratory conditions may also be observed in the field. With regard to oxides of N, the evidence includes reported effects on plant 31

photosynthesis and growth resulting from multiday exposures of six or more hours per day to NO<sub>2</sub> concentrations above 0.1 ppm. Effects occur at much lower exposures to HNO<sub>3</sub>. Laboratory and field studies report effects that include effects on tree foliage at 50 ppb ( $\sim$ 75 µg/m<sup>3</sup>) HNO<sub>3</sub> in controlled exposures and on survival of several lichen species in the Los Angeles basin during

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- 1 the 1980s. The studies vary with regard to their limitations; field studies are limited with regard
- 2 to identification of threshold exposures for the reported effects and uncertainties associated with
- 3 controlled experiments include whether the conditions under which the observed effects occur
- 4 would be expected in the field. Regardless, the elevated concentrations of NO<sub>2</sub> and HNO<sub>3</sub> in the
- 5 Los Angeles area in the 1970s-90s is well documented. For example, concentrations of HNO<sub>3</sub>
- 6 reported in forested areas of California in the 1980s ranged up to 33 ug/m<sup>3</sup>, and annual average
- 7 NO<sub>2</sub> concentrations in the Los Angeles area ranged from 0.078 ppm in 1979 to 0.053 ppm in the
- 8 early 1990s (section 5.1.2). Ambient air concentrations of HNO<sub>3</sub> in the Los Angeles metropolitan
- 9 area have declined markedly, as can be seen from Figure 2-40 (in section 2.5.4), which compares
- 10 concentrations at CASTNET monitoring sites between 2019 and 1996.
- 11 5.5.3.2 Deposition and Risks to Trees

#### 12 Soil Acidification Analyses and Risk to Trees

Steady-state modeling analysis performed in the 2009 REA estimated annual amounts of 13 acid deposition at or below which one of three BC:AL targets would be met in a 24-state area in 14 which the acid-sensitive species, red spruce and sugar maple, occur. While the two least 15 16 restrictive targets (BC:Al of 0.6 and 1) differed by less than a factor of two, the two most restrictive targets (BC:Al of 1 and 10) differed by a factor of 10. A range of acid deposition was 17 estimated for each of the three targets. For a BC:Al target of 0.6, the range was 1237-2009 eq/ha-18 yr; for a BC:Al target of 1, the range was 892-1481 eq/ha-yr; and for a BC:Al target of 10, the 19 20 range was 487-910 eq/ha-yr. Estimates of total S and N deposition in regions of the U.S. for the 2019-2021 period appear to meet all but the most restrictive of these targets (e.g., section 2.5.3 21

- above; ISA, Appendix 2, sections 2.6 and 2.7).
- 23 Uncertainties associated with these analyses include those associated with the limited
- 24 dataset of laboratory-generated data on which the BC:Al targets are based. These data are
- 25 derived from an array of studies of tree seedlings in artificial substrates and responses ranging
- from changes in plant tissue components to changes in biomass. In addition to the uncertainty
- associated with the basis for the BC:Al targets, there are uncertainties in the steady-state
- 28 modeling parameters, most prominently those related to base cation weathering and acid-
- 29 neutralizing capacity (2009 REA, section 4.3.9). As discussed in section 5.4.2.1 above, more
- 30 recent publications have employed a new approach to estimating these parameters, including the
- 31 weathering parameter, with reduced uncertainty. For the Pennsylvania study area where this was
- tested, a greater buffering capacity was estimated, and for a larger study area of the Northeast,
- the deposition estimates for the BC:Al target of 10 were slightly higher than those for the 2009
- REA (Phelan et al., 2014; Duarte et al., 2013).

#### 1 Tree Growth and Survival in Experimental Addition Studies

2 Experimental addition studies of S, or S plus N, with additions greater than 20 kg/ha/yr, have been performed in eastern locations and focused on a small set of species, including sugar 3 maple, aspen, white spruce, yellow poplar, black cherry; these studies generally have not 4 reported growth effects (Appendix 5B, section 5B.3.1). A study involving both S and N additions 5 6 greater than 20 kg/ha-yr for each substance reported increased growth rate for sugar maple but 7 not for the second species (Bethers et al., 2009), while another study of similar dosing of S and N reported reduced growth in three species after 10 years that resolved in two of the species after 8 9 22 years (Jensen et al., 2014). In both situations background deposition contributions were also appreciable (Appendix 5B, Table 5B-1). 10

11 Uncertainties associated with these analyses include the extent to which the studies reflect steady-state conditions. Given the variability in the durations across these studies and the 12 relatively shortness for some (e.g., less than five years), it might be expected that steady-state 13 14 conditions have not been reached, such that the S/N loading is within the buffering capacity of 15 the soils. A related limitation of some of these studies is the lack of information regarding 16 historic deposition at the study locations that might inform an understanding of the prior issue. 17 However, many of the studies have assessed soil characteristics and soil acidification indicators, which also informs this issue. 18

With regard to N addition, the available studies have reported mixed results for growth
and survival for several eastern species including oaks, spruce, maples and pines (Table 5B-1;
Magill et al 2004; McNulty et al 2004; Pregitzer et al 2008; and Wallace et al 2007). It is not
clear the extent to which such findings may be influenced by species-specific sensitivities or
soils and trees already impacted by historic deposition, or other environmental factors.
Uncertainties for N addition experiments and interpretation of their results include this
complexity, as well as the uncertainties identified above for S or S+N addition studies.

#### 26 *Observational/Gradient Studies of Tree Growth/ Survival*

With regard to S deposition, the two large studies that analyzed growth and/or survival measurements in tree species at sites in the eastern U.S. or across the country report negative associations of tree survival and growth with the S deposition metric for nearly half the species individually and negative associations of tree survival for 9 of the 10 species' functional type groupings (Dietze and Moorcroft, 2011; Horn et al., 2018<sup>11</sup>). Interestingly, survival for the same 9 species groups was also negatively associated with long-term average ozone (Dietze and

33 Moorcroft, 2011).

<sup>&</sup>lt;sup>11</sup> The study by Horn et al. (2018) constrained the S analyses to preclude a positive association with S.

• The full range of average  $SO_4^2$  deposition estimated for the 1994-2005 time period 1 assessed by Dietze and Moorcroft (2011) for the eastern U.S. study area was 4 to 30 kg S 2  $ha^{-1}yr^{-1}$ . 3 • Median S deposition (2000-13) estimated at sites (measurement interval average 4 [occurring within 2000-13]) of nonwestern species with negative associations with 5 growth or survival ranged from 5 to 12 kg S ha<sup>-1</sup>yr<sup>-1</sup>, with few exceptions (Horn et al., 6 7 2018). 8 The S deposition metrics for the two studies were mean annual average deposition 9 estimates for total S or sulfate (wet deposition) during different, but overlapping, time periods of roughly 10-year durations. Additionally, S deposition in the U.S. across the full period of these 10 11 studies (1994-2013) generally exhibited a consistent pattern of appreciable declines. Further, the study plots, particularly in the eastern U.S., have experienced decades of much higher S 12 deposition in the past. The extent to which the differences in growth or survival across sites with 13 different deposition estimates are influenced by to historically higher deposition (e.g., versus the 14 15 magnitude of the average over the measurement interval) is unknown. There are few available studies describing recovery of historically impacted sites (e.g., ISA, section IS.4.1, IS.5.1, 16 IS.11.2). 17 18 Regarding N deposition, the three large studies that analyzed growth and/or survival measurements in tree species at sites in the northeastern or eastern U.S., or across the country, 19 20 report associations of tree survival and growth with several N deposition metrics (Dietze and 21 Moorcroft, 2011; Thomas et al., 2010; Horn et al., 2018). • Estimates of average N deposition across the full set of sites analyzed by Thomas et al. 22 (2010) in 19 states in the northeastern quadrant of the U.S. ranged from 3 to 11 kg N/ha-23 yr for the period 2000-2004. 24 • The full range of average NO<sub>3</sub> deposition estimated for the 1994-2005 time period 25 assessed by Dietze and Moorcroft (2011) for the eastern U.S. study area was 6 to 16 kg N 26  $ha^{-1}yr^{-1}$ . 27 • Median N deposition estimated (measurement interval average [falling within 2000-13]) 28 at sites of nonwestern species for which associations with growth or survival were 29 30 negative (either over full range or at median for species) ranged from 7 to 12 kg N ha<sup>-1</sup>yr<sup>-</sup> <sup>1</sup> (Horn et al., 2018). 31 32 • Median N deposition estimated (measurement interval average [within 2000-13]) at sites of nonwestern species for which associations with growth or survival were positive 33 (either over full range or at median for species) ranged from 7 to 12 kg N ha<sup>-1</sup>yr<sup>-1</sup> (Horn 34 et al., 2018). 35 The N deposition metrics for these three studies were mean annual average deposition 36 estimates for total N or nitrate (wet deposition) during different, but overlapping, time periods 37 that varied from 5 to more than 10 years and include areas that have experienced decades of 38

much higher deposition. Further, N deposition during the combined time period (1994-2013) has
changed appreciably at many sites across the country, with many areas experiencing declines and
a few areas experiencing increases in deposition of some N species and in total N deposition.

In considering what can be drawn from these studies with regard to identification of 4 deposition levels of potential concern for tree species effects, a number of uncertainties are 5 6 recognized. For example, several factors with potential influence on tree growth and survival 7 were not accounted for. For example, although ozone was analyzed in one of the three studies, 8 soil characteristics and other factors with potential to impact tree growth and survival (other than 9 climate) were not assessed, contributing uncertainty to their interpretations. Further, differences in findings for the various species (or species' groups) may relate to differences in geographic 10 11 distribution of sampling locations, which may contribute to differences in ranges of deposition history, geochemistry etc. Additionally, as noted above, the extent to which associations reflect 12 the influence of historical deposition patterns and associated impact is unknown. 13

14 As summarized in Appendix 5B, Table 5B-6, there is a general similarity in findings among the studies, particularly of Horn et al (2018) and Dietze and Moorcroft (2011), even 15 16 though the time period and estimation approach for S and N deposition differ. Given the role of deposition in causing soil conditions that affect tree growth and survival, and a general similarity 17 18 of spatial variation of recent deposition to historic deposition, an uncertainty associated with quantitative interpretation of these studies is the extent to which the similarity in the two studies' 19 20 finding may indicate the two different metrics to both be reflecting geographic variation in impacts stemming from historic deposition. Although the spatial patterns are somewhat similar, 21 22 the magnitudes of S and N deposition in the U.S. has changed appreciably over the time period covered by these studies (e.g., Appendix 5B, Figures 5B-9 through 5B-12). The appreciable 23 24 differences in magnitude across the time periods also contribute uncertainty to interpretations 25 related to specific magnitudes of deposition associated with patterns of tree growth and survival.

#### 26 5.5.3.3 Deposition Studies of Herbs, Shrubs and Lichens

The available studies that may inform our understanding of exposure conditions, 27 28 including N deposition levels, of potential risk to herb, shrub and lichen communities include 29 observational or gradient studies and experimental addition conducted in different parts of the 30 U.S. Among the studies of plant communities are observational studies of herbaceous species richness at sites in a multi-state study area and of grassland or coastal sage scrub communities in 31 32 southern California, and experimental addition experiments in several western herb or shrub ecosystems. The experimental addition studies indicate effects on community composition 33 34 associated with annual N additions of 10 kg N/ha-yr (in the context of background deposition on 35 the order of 6 kg N/ha-yr) and higher (section 5.4.3.1 above). Experiments involving additions of 5 kg N/ha-yr variously reported no response or increased cover for one species (in context of
background deposition estimated at 5 kg N/ha-yr). The landscape-level analysis of coastal sage
scrub community history in southern California observed a greater likelihood of recovery of sites
with relatively low invasion of exotic invasive grasses when the N deposition metric level was
below 11 kg N/ha-yr. Lastly, the multi-state analysis of herbaceous species richness reported a
negative association with N deposition metric values above 8.7 kg N/ha-yr at open-canopy sites
and above 11.6 kg N/ha-yr at forest sites with acidic soil pH at or above 4.5.

8 Limitations and associated uncertainties vary between the two types of studies 9 (experimental addition and observational). Both are limited with regard to consideration of the impacts of long-term deposition. While there are some experimental addition studies lasting 10 more than 20 years, many are for fewer than 10 years. Additionally, such studies are necessarily 11 limited with regard to the number and diversity of species and ecosystems that can be analyzed. 12 13 In the case of observational studies, the many decades-long history of S and N deposition, as 14 well as elevated levels of airborne pollutants, including ozone and nitrogen oxides, in the U.S. is 15 their backdrop, and its influence on associations observed with more recent deposition metrics is generally unaccounted for. Further, given the very nature of observational studies as occurring in 16 real time, there is uncertainty associated with characterization, including quantification, of the 17 particular exposure conditions that may be eliciting patterns of ecosystem metrics observed. 18

19 The few studies of lichen species diversity and deposition-related metrics, while 20 contributing to the evidence that relates deposition, including acidic deposition in eastern locations, to relative abundance of different lichen species, are more limited with regard to the 21 22 extent that they inform an understanding of specific exposure conditions in terms of deposition 23 levels that may be of concern. As summarized in section 5.4.3.2 above, a number of factors limit 24 such interpretations of the currently available studies. These factors include uncertainties related 25 to the methods employed for utilizing estimates of N deposition, the potential role of other unaccounted-for environmental factors (including ozone, SO<sub>2</sub>, S deposition and historical air 26 27 quality and associated deposition), and uncertainty concerning the independence of any effect of deposition levels from residual effects of past patterns of deposition. We additionally note the 28 29 summary in section 5.5.3.1 above, of information on exposure conditions associated with lichen

30 species effects of oxides of N such as HNO<sub>3</sub>.
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# 6 RELATIONSHIPS OF DEPOSITION TO AIR QUALITY METRICS

## 3 6.1 OVERVIEW

To address the framing questions that guide the scope of this review, this section focuses on characterizing the relationship between deposition of S and N compounds and air quality metrics for S oxides, N oxides and PM. This characterization is a key aspect of the approach taken in this Policy Assessment (PA) for assessing deposition-related effects and the adequacy of the current secondary standards, as summarized in section 3.2 above (Figure 6-1).



10

Figure 6-1. General approach for assessing the currently available information with
 regard to consideration of protection provided for deposition-related
 ecological effects on the public welfare.

## 14 6.2 RELATING AIR QUALITY TO ECOSYSTEM DEPOSITION

- 15 While many of the ecological effects examined in this review are associated with
- 16 deposition of S and N, the NAAQS are set in terms of an ambient atmospheric concentrations.
- 17 Therefore, an important part of this review is to quantify the relationship between air
- 18 concentration and deposition. The goal of this section is to examine the relationship between air
- 19 concentrations and atmospheric deposition of S and N. Understanding more about this
- 20 relationship can then help inform how changes in air concentrations, and the emissions from
- 21 which they result, can lead to changes in the amounts of S and N deposited. This understanding

1 can then help inform decisions on the best air quality metric(s) for a standard that protects

2 against N and S deposition-related effects.

3 Atmospheric deposition of a pollutant occurs when a pollutant is transferred from the 4 atmosphere to the earth's surface through dry deposition (settling onto the surface through direct 5 contact) or wet deposition (settling onto the surface in rain, snow, or fog). There are a variety of 6 factors that determine how much of the pollutant is deposited. For example, the rate at which a 7 pollutant dry deposits (i.e., the dry deposition velocity) depends on the physical properties of the 8 chemical compound, meteorological conditions, and the surface properties. Similarly, the rate of 9 wet deposition is influenced by the physical properties of the pollutant, the precipitation rate, and 10 the vertical distribution of the pollutant in the atmosphere.

11 For dry deposition, the physical properties of a chemical compound can be especially 12 important in determining its deposition velocity and can vary as the nitrogen and sulfur 13 containing compounds change in the atmosphere. For example, NO<sub>2</sub> can oxidize to form nitric 14 acid (HNO<sub>3</sub>), which has a much higher dry deposition velocity than NO<sub>2</sub>. However, HNO<sub>3</sub> can 15 also partition into the particle phase in the presence of ammonia to form ammonium nitrate 16 (NH<sub>4</sub>NO<sub>3</sub>). Fine particles, such as PM<sub>2.5</sub>, have a much slower dry deposition velocity and remain 17 in the atmosphere longer. On the other hand, HNO<sub>3</sub> can also absorb onto larger, coarse particles, 18 whose dry deposition velocity is faster than smaller PM<sub>2.5</sub>. Thus, as the chemical and physical 19 forms of nitrogen and sulfur vary in the atmosphere, it leads to differences in the rate of 20 deposition, and causes variability in the relationship between total air concentrations and 21 atmospheric deposition. Furthermore, the dry deposition velocity is influenced by meteorological 22 conditions and their interaction with the deposition surface properties. Meteorological factors 23 such as wind speed, humidity, atmospheric stability, and temperature all affect the rate of settling 24 for particles and gases. There are also micrometeorological parameters that have an impact on 25 dry deposition of particles when they interact with surface features, such as friction velocity, 26 roughness height, and surface wetness (ISA, Appendix 2, section 2.5.2; Wesley, 2007). 27 For wet deposition, the chemical form plays a minor role, and the amount of nitrogen and 28 sulfur transferred to cloud water and falling precipitation is largely driven by the air 29 concentration. However, the vertical distribution of the pollutant is important. The air 30 concentration for the NAAQS has historically been measured near ground level where the health 31 and ecological effects occur. Atmospheric nitrogen and sulfur near the ground can settle onto 32 leaves, soils, buildings, and other surfaces by dry deposition. Sulfur and nitrogen higher in the 33 troposphere are scavenged by clouds and falling precipitation via wet deposition. While dry 34 deposition is directly related to the ground-level concentration, wet deposition is affected by 35 concentrations throughout the troposphere.

6-2

1 For ground-level emission sources, much of the nitrogen and sulfur is near the surface 2 and most of the deposition can be attributed to dry deposition. Further from emission sources, 3 pollutants become well-mixed in the atmosphere, and wet deposition can play a larger role. The 4 frequency of precipitation is also important. For example, desert areas receive very little 5 precipitation and hence contribution from wet deposition is low. Much of the western U.S. has 6 drought years that result in very low wet deposition amounts, followed by rainy years with high 7 wet deposition. The eastern U.S. has less interannual variability in rainfall. The frequency of 8 precipitation affects the relative contributions of wet and dry deposition and therefore can cause 9 variability in the relationship between ground-level air concentrations and deposition.

10 The PA in the last review introduced the Transference Ratio, defined as the ratio of 11 deposition to air concentration (2011 PA, section 7.2.3). This was calculated from annual 12 average values and spatially averaged over eco-regions that spanned distances on the order of 13 10,000 km<sup>2</sup>. While generally capturing the average relationship between air concentrations and 14 atmospheric deposition over larger areas of the country, the Transference Ratio approach has 15 some important uncertainties. For example, the transference ratio approach does not capture the 16 spatial variability across an area due to the proximity to sources, chemical composition, 17 frequency of precipitation, and vertical distribution of nitrogen and sulfur (ISA, Appendix 2, 18 section 2.5.2.4). Furthermore, the results of the approach are influenced by which air quality 19 model is used in the analyses. Studies completed since the previous review have examined how 20 the use of different models to calculate concentration and deposition can yield very different 21 estimates of the transference ratio, despite having comparable error statistics when compared to 22 measurements of air concentrations and wet deposition (ISA, Appendix 2, section 2.5.2.4). 23 This Policy Assessment recognizes these limitations, and as described in section 2, also 24 recognizes that emissions, air concentrations and deposition, have declined for sulfur and 25 oxidized nitrogen in recent years. The evolution of this trend is an opportunity to observe the 26 relationship of the change in deposition due to a change in emissions and air concentrations 27 using ongoing air concentration and wet deposition measurements. This assessment examines the 28 historical record of observations, multi-decadal CMAQ simulations, and merged model-29 measurement TDEP data to assess the relationship between air concentration of a specific 30 compound or combination of compounds and estimates of N and S deposition in specific 31 locations. After examining those relationships, this section then looks at the recent and historical 32 relationships between air concentrations of S and N and estimates of S and N deposition by

33 TDEP across the U.S.

#### 1 6.2.1 Class I Areas - Collocated Site Analyses

2 In this first set of analyses, the focus is on understanding more about the deposition of S 3 and N in remote areas that are further away from most emission sources of S and N, as well as 4 from most SO<sub>2</sub>, NO<sub>2</sub> and PM<sub>2.5</sub> FRM/FEM monitors. These areas tend to be of particular interest 5 for ecological and legal reasons, as well. Class I areas have some special federal protections 6 (e.g., focus of efforts to reduce regional haze).<sup>1</sup> For these analyses, this section analyzes 7 historical trends from measurements, CMAQ simulations, and model-measurement fusion data 8 (i.e., TDEP) to identify which S and N-related compounds are most closely related to S and N 9 deposition in these rural areas. Additionally, noting the many factors that can lead to variability 10 in estimated deposition, including frequency of precipitation, and micrometeorological factors 11 relevant to the dry deposition velocity, the analyses focus on multiple years of data to better 12 assess more typical relationships. Data for deposition and air concentration are from both 13 observations and model simulations. The air concentrations are the annual average 14 concentrations. The deposition values are the sum of total deposition (wet + dry) for the same 15 year-long period. However, when assessing deposition estimates in this part of the analysis, the 16 assessment (i.e., section 6.2.1.1) relies on wet deposition as a proxy for total deposition since dry 17 deposition is not routinely measured. 18 The set of Class I areas with co-located CASTNET monitoring stations, chemically-19 speciated PM<sub>2.5</sub> from the IMPROVE network, and NADP/NTN wet deposition monitors have 20 been identified and listed in Table 6-1 and shown in the map in Figure 6-2. There are 27 areas 21 with co-located data. The wet, dry, and total deposition are TDEP estimates, and since these data 22 are at monitoring locations, the results are largely informed by the measured values. Figure 6-3 23 shows the range of wet and dry deposition levels across these 27 areas for the 2017-2019 period. 24 For these locations, in recent years, N deposition tends to be much greater than S deposition, 25 likely due to the fact that most of these locations are in the western U.S. and distant from S 26 sources, which are principally located in the eastern U.S. S deposition has also declined more

- 27 than N deposition over the last few decades (section 2). For nitrogen, dry deposition contributes
- 28 57% and wet deposition contributes 43%. The annual total deposition from 2017 2019 for
- sulfur deposition is 60% wet deposition and 40% dry deposition.
- 30

<sup>&</sup>lt;sup>1</sup> Areas designated as Class I receive special protection status under the Clean Air Act (CAA), and include all international parks, national wilderness areas which exceed 5,000 acres in size, national memorial parks which exceed 5,000 acres in size, and national parks which exceed 6,000 acres in size, provided the park or wilderness area was in existence on August 7, 1977. Other areas may also be Class I if designated as Class I consistent with the CAA.

Class I Area name	CASTNET	NADP	IMPROVE
Acadia	ACA416	ME98	ACAD1
Big Bend	BBE401	TX04	BIBE1
Canyonlands	CAN407	UT09	CANY1
Chiricahua	CHA467	AZ98	CHIR1
Death Valley	DEV412	CA95	DEVA1
Dinosaur National Monument	DIN431	CO15	DINO1
Everglades	EVE419	FL11	EVER1
Glacier	GLR468	MT05	GLAC1
Great Basin	GRB411	NV05	GRBA1
Grand Canyon	GRC474	AZ03	GRCA2
Great Smokey Mountains	GRS420	TN11	GRSM1
Joshua Tree	JOT403	CA67	JOSH1
Mt. Lassen	LAV410	CA96	LAVO1
Mammoth Cave	MAC426	KY10	MACA1
Mesa Verde	MEV405	CO99	MEVE1
Cascades	NCS415	WA19	NOCA1
Olympic	OLY421	WA14	OLYM1
Petrified Forest	PET427	AZ97	PEFO1
Pinnacles	PIN414	CA66	PINN1
Rocky Mountain	ROM406	CO19	ROMO1
Sequoia	SEK430	CA75	SEQU1
Shenandoah	SHN418	VA28	SHEN1
Theodore Roosevelt	THR422	ND00	THRO1
Voyageurs	VOY413	MN32	VOYA2
Wind Cave	WNC429	SD04	WICA1
Yellowstone	YEL408	WY08	YELL2
Yosemite	YOS404	CA99	YOSE1

1Table 6-1.Co-located CASTNET, NADP/NTN, and IMPROVE monitoring stations used2in this analysis of air concentration and deposition.



Figure 6-2. Locations of co-located CASTNET, NADP/NTN, and IMPROVE monitoring sites, denoted by CASTNET site identifier. The NADP/NTN and IMPROVE station identifiers are listed in Table 6-1.



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Figure 6-3. Dry and wet deposition of nitrogen and sulfur (2017-2019 annual average),
 for locations listed in Table 6-1.

1 In the following three subsections (6.2.1.1, 6.2.1.2, 6.2.1.3), the analyses focus on assessing 2 relationships between: (1) wet deposition measurements and air concentration measurements; (2) 3 CMAQ simulations, to understand the air concentration and total deposition relationship from 4 the perspective of a model that reflects known physical and chemical processes; and (3) 5 measured air concentrations and the total deposition estimated by TDEP at the same location. 6 These sets of measured and predicted variables are compared using linear regression which 7 allows a more detailed assessment of the uncertainty and variability. There are several ways to 8 assess how well one variable relates to the other, such as by calculating the correlation (r), the 9 coefficient of determination  $(r^2)$ , the distribution of the residuals, and the uncertainty in the 10 assessment of the slope.

11

#### 6.2.1.1 Evidence from Observations of Air Concentrations and Wet Deposition

This section assesses the relationships between wet deposition measurements and air concentration measurements. Wet deposition is measured by the NADP/NTN network. CASTNET measures particle sulfate and nitrate, gas phase SO<sub>2</sub>, and gas phase HNO<sub>3</sub>. The IMPROVE network measures total PM<sub>2.5</sub> and the sulfate and nitrate components of PM<sub>2.5</sub>. These three types of monitors are collocated at 27 different sites listed in Table 6-1 and shown on the map in Figure 6-2.

The comparison between annual average PM2.5 air concentration measurements from 18 19 IMPROVE and annual total wet deposition measurements from NADP/NTN is shown in Figure 20 6-3. For this subset of 27 Class I areas, the data indicates that wet S deposition is most highly correlated with  $SO_4^{2-}$  (r = 0.87). The correlation between wet S deposition and total PM<sub>2.5</sub> is less 21 22 (r = 0.73) and, as expected, there is little correlation between wet S deposition and NO<sub>3</sub><sup>-</sup> (r = 0.73)23 0.14). This figure also shows that the combined S and N wet deposition at these sites is very 24 highly correlated (r = 0.99) with wet S deposition. Turning attention to how wet N deposition 25 measurements relate to pollutant concentration data from IMPROVE, there is some moderate positive correlation with observed  $SO_4^{2-}$  (r = 0.70) and total PM<sub>2.5</sub> (r = 0.64), but not with NO<sub>3</sub><sup>-</sup> (r 26 27 = 0.28). The scatterplot for this pairing suggests that there are many location/years where the 28 annual average NO<sub>3</sub><sup>-</sup> data are relatively high (e.g., 1-2  $\mu$ g/m<sup>3</sup>) but wet N deposition remains 29 relatively low. The low correlation between nitrate PM<sub>2.5</sub> and N deposition may be due to uncertainty in the nitrate PM2.5 measurement, which is larger than sulfate PM2.5 uncertainty, or to 30 a larger role for ammonium  $PM_{2.5}$  in N deposition (NH<sub>4</sub><sup>+</sup> not measured by IMPROVE). An 31 32 additional explanation is that nitric acid also contributes to N deposition and is an additional 33 source of variability not captured by PM nitrate.

To address the contribution of nitric acid, CASTNET air concentration measurements of total sulfur and total nitrate ( $HNO_3 + NO_3^{-}$ ) were compared to wet deposition measurements

6-7

- 1 from NADP/NTN, as shown in Figure 6-5. As in the IMPROVE case, there is strong correlation
- 2 between S wet deposition and concentrations of total sulfate (r = 0.88) but again comparatively
- 3 weaker correlation between N wet deposition and measured concentrations of total nitrate (r =
- 4 0.38). Comparing the x-axes for IMPROVE  $NO_3^-$  (Figure 6-4) and CASTNET total nitrate
- 5 (Figure 6-5) shows that CASTNET total nitrate spans a factor of two larger range than
- 6 IMPROVE NO<sub>3</sub>, so we conclude that most of the total nitrate is in the form of nitric acid at these
- 7 sites. This is captured in Figure 6-5 which shows that, for these sites, the composition of S
- 8 concentration between  $SO_2$  and  $SO_4^{2-}$  is more evenly split. This is an artifact of most of these 27
- 9 Class I areas being located in the western U.S. where S is generally low and concentrations of
- 10 SO<sub>2</sub> and SO<sub>4</sub><sup>2-</sup> tend to be similar. (In other parts of the country SO<sub>2</sub> tends to be higher near
- 11 emissions sources of SO<sub>2</sub>, with a greater chance of oxidation to  $SO_4^{2-}$  higher at farther distances.)



Figure 6-4. Scatter plot matrix of annual average wet deposition measurements from NADP/NTN (5 pollutants, units: kg/ha-yr) versus annual average concentrations from IMPROVE (3 pollutants, units: μg/m3) for 27 Class 1 areas from 1988-2018. A histogram of each deposition or concentration variable is shown in a diagonal running from the top left to lower right. Below that diagonal are scatter plots for each pair of variables. Above that diagonal are the correlations between pairs of variables. (Note for this plot and all subsequent matrix plots: the x- and y- axes scales are shown on the left and right sides of the plot for rows, and at the top and bottom of the plot for columns.)



1 2 3 4 5 6 7 8

diagonal are the correlations between pairs of variables.

areas from 1988-2018. A histogram of each deposition or concentration

variable is shown in a diagonal running from the top left to lower right. Below that diagonal are scatter plots for each pair of variables. Above that





Figure 6-6.

3 4 5

Histograms of the ratios of the gas phase SO<sub>2</sub> to particle SO<sub>4</sub><sup>2-</sup> (left) and the gas phase HNO<sub>3</sub> to particle NO<sub>3</sub><sup>-</sup> (right) in CASTNET data. Each ratio is calculated as the annual average concentration (2000-2019), converted to moles of N or moles of S, for the 27 sites listed in Table 6-1.

6 One possible explanation for why particle sulfate is more strongly correlated with sulfur 7 wet deposition while particle nitrate has weaker correlation with nitrogen wet deposition is the 8 chemical properties of these compounds. Particle sulfate can be formed in clouds, it has 9 relatively low spatial variability, and SO<sub>2</sub>, while a minor contributor to wet deposition, is highly correlated with particle sulfate (r = 0.91 at CASTNET sites, not shown in figures). Particle 10 nitrate concentrations have larger spatial variability as the partitioning between gas-phase nitric 11 12 acid and particle nitrate is controlled by temperature, relative humidity, and the availability of 13 cations such as ammonium. In the CASTNET measurements, the correlation between nitric acid 14 and particle nitrate is lower (r = 0.47, not shown in figures) and at CASTNET sites, nitric acid is 15 much more abundant than nitrate PM (Figure 6-6) although this interpretation should be 16 tempered due to uncertainties in the CASTNET measurement technique that make it difficult to 17 differentiate nitrate PM and nitric acid. 18 The evidence from observations of air concentrations and wet deposition (as a proxy for 19 total deposition) at 27 U.S. sites with collocated measurements of air quality and deposition

20 suggest that particle sulfate is strongly correlated with wet S deposition, but that particle nitrate

6-11

and total nitrate ( $HNO_3 + NO_3^{-}$ ) are not as strongly correlated with wet N deposition. Both wet deposition of S and N are moderately correlated with total measured  $PM_{2.5}$ .

3

#### 6.2.1.2 Evidence from Chemical Transport Modeling

4 Since dry deposition flux is not routinely measured, models are often used to examine the 5 relationship between air concentration and total deposition. The Community Multiscale Air 6 Quality Modeling System (CMAQ) is a numerical air quality model that relies on scientific first 7 principles to predict the concentration of airborne gases and particles, and the deposition of these 8 pollutants back to Earth's surface. The results of a 21-year CMAQ simulation have been made 9 available, as described in Zhang et al. (2018). We utilize these model simulations to further 10 analyze relationships between air concentrations and deposition of S- and N-related compounds 11 as part of this review. 12 Figures 6-7 and 6-8 show spatial maps of the annual average  $SO_X$  and  $NO_Y$ 13 concentrations (left panel), total S and N deposition (middle panel), and the 14 deposition/concentration ratio for oxidized sulfur and total nitrogen (right panel). For S oxides 15 (Figure 6-7), most of the U.S. exhibits deposition/concentration ratios of 0.5 to 3, most notably in 16 areas where local and regional sources of SO<sub>2</sub> are prevalent. However, as an airmass moves 17 further away from emissions sources, the more rapidly depositing compounds are removed, and 18 pollutants are diluted by being mixed vertically in the atmosphere. In these locations, higher 19 deposition-to-concentration ratios for S oxides are modeled. Examples include parts of the 20 northeastern U.S. and at high elevation sites in the western U.S. These areas are farther away 21 from sources and ground-level air concentrations are low; however, sulfate can be transported in 22 clouds and deposited by falling rain, leading to a high level of deposition, relative to the ground-23 level air concentration. For N, the spatial patterns are similar, however the ratios are slightly 24 lower over most of the U.S. (i.e., ratios range from 0.5 to 2). Again, while the spatial distribution 25 of the concentration and deposition suggests that there is a strong correspondence, the ratio of the 26 two terms can vary spatially.





Figure 6-8. Annual average concentration (μg/m<sup>3</sup>), deposition (kg/ha-yr), and the deposition/concentration ratio for nitrogen compounds, as estimated using a 21-year (1990-2010) CMAQ simulation.

6-14

1

Because there is evidence of variability in the deposition and concentration relationship, it is important to rigorously assess potential deposition predictors. In order to compare the CMAQ model results against the previous analysis of the concentration and deposition relationships at 27 monitoring sites with collocated data, the EPA evaluated data from the grid cells representing those 27 Class I areas. The matrix scatterplots of these results are displayed in Figure 6-9.

7 Starting with a comparison of the wet deposition only results, it can be noted that the air 8 quality modeling data indicates strong correlation between total sulfate and wet S deposition (r = 9 0.90). In the model data, unlike what was observed from the measurement data, there is also 10 relatively strong correlation between total nitrate and wet N deposition (r = 0.76). For both S and 11 N, the correlation of wet deposition with total  $PM_{2.5}$  is slightly greater in the model data (r = 12 0.81, for both pollutants) than in the observed data (r = 0.73 and r = 0.64, respectively). 13 In Figure 6-9, it can be seen that total S and N deposition in the model output is strongly 14 correlated with wet deposition of S and N at these 27 sites (r = 0.98 and r = 0.86, respectively), 15 confirming our earlier assumption that most of the deposition at these locations likely occurs 16 through wet deposition. However, the advantage of the simulation data is that we can also 17 evaluate the relationships between concentration data and total deposition (wet + dry). As 18 expected, and consistent with previous results, total S deposition is strongly correlated with total 19 sulfur (r = 0.95) in CMAQ at these locations. Interestingly, the model data also show a strong 20 correlation between total N deposition and total nitrate (r = 0.94). As the modeling data includes 21 ammonium in total nitrate, unlike the IMPROVE and CASTNET data, this suggests that the 22 weaker correlations in the observed data may have been driven by incomplete measurements of 23 the total nitrate and/or uncertainties in the measurement data. For PM<sub>2.5</sub>, the data suggests strong 24 correlation in the model results with both total S deposition (r = 0.85) and total N deposition (r =25 0.91).



2 Scatter plot matrix of annual average CMAQ-simulated total deposition (4 Figure 6-9. 3 pollutants, units: kg/ha-yr) versus annual average CMAQ-simulated 4 concentrations (3 pollutants, units: µg/m3) for 27 Class 1 areas from 1988-5 2018. A histogram of each deposition or concentration variable is shown in a 6 diagonal running from the top left to lower right. Below that diagonal are 7 scatter plots for each pair of variables. Above that diagonal are the 8 correlations between pairs of variables. Each data point is the annual 9 average air concentration or annual total deposition from a 21-year CMAQ 10 simulation (1990-2010). The CMAQ model is sampled at 27 locations within Class I Areas with collocated CASTNET and NADP/NTN samplers listed in 11 12 Table 6-1.

1

#### 6.2.1.3 Evidence from Model-measurement Fusion

The TDEP approach described in section 2.5 estimates total deposition using a combination of measurements from NADP/NTN and CASTNET fused with CMAQ simulation results. This section compares the TDEP estimates of deposition with air concentration measurements of PM<sub>2.5</sub>, total nitrogen, and total sulfur at the sites listed in Table 6-1.

6 Starting again with total S deposition, the comparison of IMPROVE (Figure 6-10) and

7 CASTNET (Figure 6-11) air quality concentrations and TDEP deposition data suggest that S

8 deposition is again reasonably well correlated with total sulfate, both in the IMPROVE data (r =

9 0.79) and CASTNET data (r = 0.87). For N deposition, the TDEP comparisons confirm the

10 observed wet deposition comparisons in section 6.2.1.1. That is, IMPROVE nitrate data is only

11 weakly correlated with total N deposition (r = 0.31). The strength of the relationship is improved

12 when total N deposition is compared again CASTNET total nitrate (i.e., with the inclusion of

13 nitric acid) but is still more weakly correlated (r = 0.65) than what is seen for sulfur. The

14 correlation between measured  $PM_{2.5}$  and TDEP deposition estimates (r = 0.78 for S, r = 0.73 for

15 N) is just slightly higher than what was determined when evaluating the wet deposition

16 observations, and just slightly less than what was noted from the CMAQ data. All three

17 evaluation approaches showed similar correlation between N and S deposition and PM<sub>2.5</sub> data

18 (ranges from r = 0.64 to r = 0.81).



Figure 6-10. Scatter plot matrix of annual average TDEP deposition (3 pollutants, units: kg/ha-yr) versus annual average IMPROVE concentrations (5 pollutants, units: μg/m3) for 27 Class 1 areas with collocated IMPROVE and NADP/NTN from 1988-2018. A histogram of each deposition or concentration variable is shown in a diagonal running from the top left to lower right. Below that diagonal are scatter plots for each pair of variables. Above that diagonal are the correlations between pairs of variables.



1 2 3 4 5 6 7 8 9 10

Figure 6-11. Scatter plot matrix of annual average TDEP deposition (3 pollutants, units: kg/ha-yr) versus annual average CASTNET concentrations (5 pollutants, units: μg/m3) for 27 Class 1 areas with collocated CASTNET and NADP/NTN from 1988-2018. A histogram of each deposition or concentration variable is shown in a diagonal running from the top left to lower right. Below that diagonal are scatter plots for each pair of variables. Above that diagonal are the correlations between pairs of variables.

9 Figure 6-12 shows the relationship between S deposition and IMPROVE (PM<sub>2.5</sub> and 10 sulfate) and CASTNET (total sulfate) air concentration in more detail and helps evaluate how 11 this relationship may be changing with time. In this plot, which considers 3-year averages over 12 the 2002-2019 period, the colored dots represent more recent data (i.e., blue dots: 2014-2016 and 13 red dots: 2017-2019). It is quickly evident that air quality concentrations are lower at these 27

14 sites in recent periods relative to the past. And as noted above, IMPROVE PM<sub>2.5</sub> has some

1 correspondence with S deposition but there are a number of outliers where the PM<sub>2.5</sub>

2 concentration is high, but the S deposition is very low, especially in more recent years (colors).

- 3 These are likely cases where the  $PM_{2.5}$  is mostly composed of compounds other than sulfate.
- 4 For nitrogen (Figure 6-13), IMPROVE PM<sub>2.5</sub>, IMPROVE inorganic N PM<sub>2.5</sub> (NO<sub>3</sub><sup>-</sup> +
- 5  $NH_4^+$ ,  $\mu g N m^{-3}$ ), and inorganic nitrogen measured at CASTNET monitoring sites (HNO<sub>3</sub> + NO<sub>3</sub><sup>-</sup>
- $6 + NH_4^+$ ,  $\mu g N m^{-3}$ ) are the most closely associated with TDEP N deposition. IMPROVE
- 7 ammonium is estimated assuming that the nitrate and sulfate are fully neutralized by ammonia. A
- 8 large ratio of organic to inorganic  $PM_{2.5}$  may challenge this approach (Silvern et al., 2017).
- 9 However, this assumption may be adequate for IMPROVE sites, which are generally in the
- 10 western U.S. where there is a smaller contribution to  $PM_{2.5}$  from biogenic emissions. IMPROVE
- 11 PM<sub>2.5</sub> has the widest prediction interval, while IMPROVE inorganic N PM<sub>2.5</sub> and total inorganic
- 12 N measured at CASTNET have similar correlations to N deposition, with the CASTNET total
- 13 inorganic N having slightly fewer outliers.
- 14

## 6.2.1.4 Conclusions

15 The above analyses focus on characterizing relationships between various chemical 16 species that are the air quality components of S and N and deposition of S and N over longer 17 time periods (e.g., annual or 3-year averages) in more rural locations by assessing various forms 18 of available information collocated (measured and estimated) at 27 sites in Class I areas. 19 Assessment of these various forms of information generally show consistency in the observed 20 relationships. For S, the analyses suggest that in more rural locations, such as those represented 21 by these 27 Class I areas, S deposition is most strongly associated with measurements of both 22 sulfate and total sulfur. There is a slightly weaker association between S deposition and PM<sub>2.5</sub> in 23 these rural locations, marked by more variability, as some percentage of the PM<sub>2.5</sub> mass is expected to be composed of compounds other than sulfate. These results suggest that S 24 25 deposition in rural areas is mostly resulting from deposition of sulfate and SO<sub>2</sub>. This is consistent 26 with our understanding of the chemical properties and physical transport of these compounds. 27 For example, we know that fine particles, such as PM<sub>2.5</sub>, have a much slower dry deposition 28 velocity and remain in the atmosphere longer. Thus, it is not surprising to see that sulfur can 29 transported as PM<sub>2.5</sub> in these rural locations. These results also suggest that IMPROVE PM<sub>2.5</sub>, 30 IMPROVE sulfate, and total sulfur measured at CASTNET all could potentially be used to 31 predict S deposition, with CASTNET S showing the strongest relationship over recent years. For 32 N, these results suggest that N deposition in these rural areas is only somewhat correlated with 33 air concentrations of nitric acid and particulate nitrate. However, the results suggest that 34 IMPROVE  $PM_{2.5}$ , IMPROVE approximated inorganic N  $PM_{2.5}$  (NO3<sup>-</sup> + NH4<sup>+</sup>), and 35 approximated inorganic nitrogen measured at CASTNET monitoring sites (HNO3 + NO3<sup>-</sup> +

1 NH4<sup>+</sup>) can be used to predict N deposition in these locations, with CASTNET N showing the

2 most consistent relationship over recent years.

3

4

11



Figure 6-12. TDEP sulfur deposition (vertical axis) and air concentration (horizontal axis)
for IMPROVE PM2.5 (left), IMPROVE SO4<sup>2-</sup> (center) and CASTNET total
sulfur (right) as three-year averages from 2002–2019. Blue dots (2014-2016)
and red dots (2017-2019) show more recent data. A black dashed line denotes
the best fit using linear regression and the grey dashed lines denote the 90%
prediction interval.



12Figure 6-13.TDEP Nitrogen deposition (vertical axis) and air concentration (horizontal13axis) for IMPROVE PM2.5 (left), IMPROVE PM2.5 inorganic nitrogen14(center), and CASTNET inorganic nitrogen (right) as three-year averages15from 2002 - 2019. Blue dots (2014-2016) and red dots (2017-2019) represent16more recent data. A black dashed line denotes the best fit using linear17regression and the grey dashed lines denote the 90% prediction interval.

#### 1 6.2.2 National-scale Sites of Influence Analyses

To broaden the geographical scope of our assessment, this section incorporates information about deposition across the U.S. and analyzes the quantitative relationships between 1) the concentrations of S and N-related compounds measured at ambient monitors used to judge attainment of the current secondary NAAQS for oxides of nitrogen, oxides of sulfur and PM and 2) the magnitude of S and N deposition.

#### 7 **6.2.2.1** Approach

8 Changes in measured concentrations of NO<sub>2</sub>, SO<sub>2</sub>, and PM at ambient monitors are an 9 indicator of the changes occurring in related sources of emissions. To better understand the 10 relationship between these measured air quality concentrations and S and N deposition in various 11 downwind locations of significance, this assessment uses the HYSPLIT air parcel trajectory 12 model to examine the transport of pollutant material from source to receptor. In this analysis, the 13 EPA utilized all NO<sub>2</sub>, SO<sub>2</sub>, and PM<sub>2.5</sub> ambient air quality monitor locations for which valid 14 design values exist (i.e., from the SLAMS network described in section 2-3), in conjunction with 15 the HYSPLIT model, to identify meteorological patterns and estimate how pollution observed at 16 certain locations (referred to here as "sites of influence") could be transported to ecoregions 17 within the U.S. For PM, the analysis focuses on assessing the PM<sub>2.5</sub> annual standard in the sites of influence analyses because most deposition will transport in the smaller size fraction (i.e., as 18 19 PM<sub>2.5</sub> rather than PM<sub>10</sub> or greater) and because an annual average standard is more relevant to 20 assessing accumulating deposition than a standard with a form set to reduce peak concentrations 21 (i.e., PM<sub>2.5</sub> 24-hour standard with its 98<sup>th</sup> percentile form). The output from this analysis was 22 then postprocessed and associated with ambient measurements of concentrations of NO<sub>2</sub>, SO<sub>2</sub>, and PM<sub>2.5</sub>, and TDEP estimates of S and N deposition for a range of years dating back to 2001. 23 24 By identifying which air quality monitors are potentially representative of the air quality that 25 leads to deposition in a particular ecoregion (see Figure 6A-1 for an example sites of influence 26 set), one can better understand the relationship between upwind ambient air concentrations and 27 downwind deposition. 28 After identifying the upwind geographic areas from which emissions potentially 29 contribute to N and S deposition in each Ecoregion III areas, the EPA analyzed air quality design 30 values within each Ecoregion's set of sites of influence to estimate a weighted-average design

- 31 value<sup>2</sup>, which we call an Ecoregion Air Quality Metric (EAQM). EAQM values were estimated
- 32 for each Ecoregion III area and for three separate pollutants: NO<sub>2</sub>, SO<sub>2</sub>, and PM<sub>2.5</sub>, and are
- 33 intended to provide a perspective of air quality levels in the upwind regions that potentially

<sup>&</sup>lt;sup>2</sup> For this, EPA calculates EAQM values for each Ecoregion by weighting the design value concentration at each monitor by the percentage of HYSPLIT trajectories estimated to be linked to the Ecoregion III area.

1 contribute to downwind deposition levels. For SO<sub>2</sub>, EPA also estimated EAQM values for SO<sub>2</sub>

- 2 using an annual average given the cumulative effect of deposition that might correspond best to a
- 3 longer averaging period.<sup>3</sup>

4 As shown in section 6.2.1, the linkage between air concentration and deposition can vary, 5 even at collocated sites. This variability can be influenced by meteorology, including frequency 6 of precipitation and micrometeorological factors relevant to the dry deposition velocity. This 7 analysis aimed to reduce biases due to meteorological variations by focusing on multiyear 8 averages of deposition. To provide information across a long time period that includes the 9 important reductions in emissions of N and S described in section 2.4, the assessment evaluates 10 data over 20 years, with a focus on the following set of years: 2001-2003, 2006-2008, 2010-11 2012, 2014-2016 and 2018-2020.

12 The methodology used to calculate the air parcel trajectories that led to the sites of 13 influence identification, as well as the methodologies used to estimate the EAQM values for each 14 Ecoregion/pollutant pair using historical air quality design value (DV) data can be found in 15 Appendix 6A. In addition, to the EAQM values, EPA also extracted the highest monitored 16 design value in an area contributing pollution to each ecoregion. The EAQM is useful in 17 assessing how well measured air quality metrics for various S and N related pollutants are 18 correlated with estimated S and N deposition. Because the EAQM is a weighted metric of 19 concentration measurements from a number of monitors, it cannot be used alone to quantify how 20 the level of a design value at one monitor would correspond to a level of deposition in one area. 21 Similarly, the same is true for the information from the other analyzed metric – the maximum 22 design value from contributing monitors. However, used together, assessment of these two 23 metrics can help inform the range of levels associated with certain air quality metrics that might 24 be used to maintain S and N deposition at or below certain levels across the U.S. For example, 25 the EAQM can be viewed as providing information about a "typical" or "average" contributing 26 design value level, with some monitors measuring higher and some measuring lower yet being 27 associated with the same deposition level. On the other hand, the maximum design value from 28 contributing monitors can be viewed as providing information on the highest design value 29 associated with a particular deposition level. As shown in the figures below, the EAQM tends to 30 be better correlated with deposition, when compared to the maximum concentration at the 31 contributing monitor, for both S and N deposition and for all measured air quality metrics. This 32 is not a surprising result given that deposition is a function of accumulated deposition over 33 several years and contributed to by pollution from multiple locations. However, the measured 34 concentration at the maximum contributing monitor does also show a relationship for most of the

<sup>&</sup>lt;sup>3</sup> An annual average SO<sub>2</sub> standard was established in 1971 but revoked in 1973 (38 FR 25678, September 14, 1973).

1 air quality metrics. Table 6-2 shows the air quality metrics that were included in this assessment

2 and shown in Figure 6-14 to 6-26.

3

Figure Number	Y-Axis Metric	Pollutant	X-Axis Metric		
6-14	Estimated 3-year average S deposition (ecoregion median)			EAQM, 3-year average	
6-15			2 <sup>nd</sup> highest 3-hr average	Maximum, 3-year average	
6-16	Number of ratios			Bins of Maximum to EAQM ratios, 3-year average	
6-17	Estimated 3-year average S deposition (ecoregion median) Number of ratios	502		EAQM, 3-year average	
6-18			Annual average	Maximum, 3-year average	
6-19				Bins of Maximum to EAQM ratios, 3-year average	
6-20	Estimated 3-year average N deposition (ecoregion median) Number of ratios	Estimated 3-year average N deposition (ecoregion median)NO2Number of ratiosNO2			EAQM, 3-year average
6-21			NO <sub>2</sub>	Annual	Maximum, 3-year average
6-22			average	Bins of Maximum to EAQM ratios, 3-year average	
6-23	Estimated 3-year average			EAQM 3-year average	
6-24	S deposition (ecoregion median)         Estimated 3-year average         N deposition (ecoregion median)         PM <sub>2.5</sub>		Annual average	Maximum, 3-year average	
6-25				EAQM, 3-year average	
6-26		PM <sub>2.5</sub>		Maximum, 3-year average	
6-27				Bins of Maximum to EAQM ratios, 3-year average	
6-28	Estimated 3-year average			EAQM 3-year average	
6-29	median)			Maximum, 3 year average	

4	Table 6-2.	Relationship of deposition (S and N) to the various air quality metric	s.
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## 6

## 6.2.2.2 SO<sub>2</sub> Results

Figure 6-14 displays a comparison of 3-year average sulfur deposition (i.e., median of
TDEP values within an ecoregion) against the 3-year EAQM for the current secondary SO<sub>2</sub>
standard (i.e., annual 2<sup>nd</sup> high of individual 3-hour SO<sub>2</sub> averages). The data are binned into five
distinct time periods as shown in the legend. The figure reaffirms the decreasing trends in
ambient SO<sub>2</sub> and S deposition discussed in section 2. Prior to the 2010-2012 period, it was not
uncommon for ecoregions to experience median S deposition values exceeding 5 kg/ha-yr. Since

- 1 the 2014-2016 period, however, no regions have experienced median S deposition above that
- 2 level. At the same time, the secondary SO<sub>2</sub> EAQM has also trended downward across the
- 3 ecoregions. There is a positive and moderately strong correlation (r = 0.75) between S deposition
- 4 in an ecoregion and the weighted design values of the current secondary SO<sub>2</sub> standard in upwind
- 5 areas potentially affecting that ecoregion.
- 6



### Figure 6-14. Scatterplot of estimated 3-year average S deposition (ecoregion median) and the weighted secondary SO<sub>2</sub> design values from contributing upwind areas for that ecoregion (EAQM) also averaged over 3 years.

11 As introduced above in section 6.2.2.1, this assessment also considered the relationship 12 between TDEP-estimated deposition in the ecoregions and the maximum design value monitored 13 anywhere within the set of sites of influence for the ecoregion over a three-year period. Figure 6-14 15 displays this comparison for S deposition and the current secondary SO<sub>2</sub> standard. Again, the 15 data are binned into the same five time periods. As can be seen by the expanded x-axis in Figure 16 6-15, the maximum secondary SO<sub>2</sub> design values can be considerably higher than the weighted averages of the EAQMs. Even for the more recent time periods, there are ecoregion-influencing 17 18 sites where the second-highest annual 3-hour SO2 values exceeds 250 ppb (highest = 386 ppb). 19 While there is a positive correlation between S deposition and this potential indicator, it is less 20 than what was observed with the EAQM (r = 0.40), suggesting this metric is somewhat less

- 1 useful in linking upwind concentrations to downwind deposition. Figure 6-16 shows the
- 2 relationship between the secondary SO<sub>2</sub> EAQM values (i.e., weighted across all contributing
- 3 monitors) and the secondary SO<sub>2</sub> DVs from the maximum contributing monitors. Most
- 4 maximum/EAQM ratios range from 2-5, although there are exceptions where the ratios can be
- 5 higher than 10. One possible cause for an exceptionally high ratio would be a situation in which
- 6 there are a number of potential contributing monitors to an ecoregion but where one of the
- 7 monitors is particularly affected by a single emissions source and consequently has a higher DV
- 8 than the other contributors. The value of the weighted EAQM approach is that it attempts to
- 9 account for the expected contribution of the outlier in this hypothetical relative to the other
- 10 contributing monitor locations, by evaluating the frequency of wind trajectories. However, as
- 11 discussed further in section 6.2.2.1, there is also some value in the maximum.



Figure 6-15. Scatterplot of estimated 3-year average S deposition (ecoregion median) and
 the secondary SO<sub>2</sub> design value over that 3-year period from the contributing
 monitor with the maximum value for each ecoregion.


Figure 6-16. Histogram of the ratio of secondary SO<sub>2</sub> design value (ppb) from the
 maximum contributing monitor for that ecoregion to the average of weighted
 secondary SO<sub>2</sub> design values (EAQM) (median = 4).

5 As noted earlier, the EPA has in the past promulgated a secondary SO<sub>2</sub> standard based on 6 an annual average. When considering deposition-related effects which are cumulative in nature, 7 there may be some advantage in linking annual average concentrations to the eventual 8 deposition. Figures 6-17, 6-18, and 6-19 repeat the analysis described above but using annual 9 average SO<sub>2</sub> metrics (EAQM and maximum) instead of the current secondary SO<sub>2</sub> standard (i.e., 10 2<sup>nd</sup> highest 3-hour maximum value). The positive correlation between an EAQM based on annual 11 average SO<sub>2</sub> concentrations and S deposition in the ecoregions is slightly stronger (r = 0.81) than 12 what was observed with the shorter-term form of the standard (r = 0.75). This suggests that 13 consideration of a longer averaging time might be an important consideration in any revised 14 NAAQS. Figure 6-17 displays the relationship. There are a subset of sites with very low 15 deposition (i.e., < 5 kg/ha-yr) where there is very little association between the upwind 16 concentrations and ecoregion deposition. This observation suggests that the relationship between 17 upwind SO<sub>2</sub> concentrations and eventual downwind deposition may break down at lower 18 deposition levels (i.e., there may be factors other than contemporaneous air quality which 19 determine the deposition amounts). However, for ecoregions where S deposition is higher (i.e., > 20 5 kg/ha-yr), there is strong correlation. Figure 6-18 shows the comparison between ecoregion S 21 deposition and maximum measured annual average SO<sub>2</sub> concentrations in potentially influencing 22 upwind areas. Again, the correlation is slightly stronger than what is seen with the current 23 secondary NAAQS (r = 0.50 vs. r = 0.40). Also, the use of the maximum observations as 24 opposed to the weighted EAQM again results in weaker associations (r = 0.50 vs. r = 0.81). 25 Finally, Figure 6-19 shows the ratios between the two terms (maximum/EAQM). The ratios

- 1 between these two terms is slightly lower when considering a longer averaging time but still
- 2 most often ranges from 2-4 and there can still be values in excess of 10.



# Figure 6-17. Scatterplot of 3-year average S deposition (ecoregion median) and the weighted annual average SO<sub>2</sub> concentrations from contributing upwind areas for that ecoregion (EAQM) also averaged over 3 years





Figure 6-18. Scatterplot of estimated 3-year average S deposition (ecoregion median) and the annual average SO<sub>2</sub> concentration over that 3-year period from the contributing monitor with the maximum value for each ecoregion.



8

Figure 6-19. Histogram of the ratio of annual average SO<sub>2</sub> concentration (ppb) averaged over a 3-year period from the contributing monitor with the maximum value for each ecoregion to the average of weighted annual average SO<sub>2</sub> design values (EAQM) over the same 3-year period.

### 6.2.2.3 NO<sub>2</sub> Results

2 Similar analyses were completed assessing the relationship between the current 3 secondary NO<sub>2</sub> standard (annual mean, level = 53 ppb). Based on the results of section 6.2.1, one 4 would expect it to be less likely that the existing NO<sub>2</sub> NAAQS would be strongly correlated with 5 N deposition (due to the multiple pathways for N deposition, including ammonia-related 6 sources). Figure 6-20 displays a comparison of 3-year average N deposition estimates (TDEP) 7 against EAQM values for annual average NO<sub>2</sub>. While the data suggest that the ecoregions with 8 higher N depositions are associated with higher EAQM values, the correlation is less strong than 9 what was seen for SO<sub>2</sub> (r = 0.58 vs. r = 0.75). However, unlike SO<sub>2</sub>, the positive association 10 appears to extend throughout the distribution of N deposition levels; that is, the correlation 11 between deposition and EAQM is similar whether N deposition values are greater than, or less 12 than, for example 10 kg/ha-yr. As was the case for SO<sub>2</sub>, Figure 6-21 illustrates that the switch to 13 consideration of the single highest NO<sub>2</sub> DV from the set of contributing monitors, as opposed to 14 a weighted EAOM value, slightly reduces the correlation between deposition and concentration (r = 0.35 vs. r = 0.58). The NO<sub>2</sub> ratios between maximum DVs and EAQM values typically 15 16 range from 1.5 to 2.5 but can be as high as 6.5.



## Figure 6-20. Scatterplot of estimated 3-year average NO<sub>2</sub>, average for 3-yr period (ppd) Figure 6-20. Scatterplot of estimated 3-year average N deposition (ecoregion median) and the weighted secondary NO<sub>2</sub> design values from contributing upwind areas for that ecoregion (EAQM) also averaged over 3 years.





4

Figure 6-21. Scatterplot of estimated 3-year average N deposition (ecoregion median) and the secondary NO<sub>2</sub> design value over that 3-year period from the contributing monitor with the maximum value for each ecoregion.



8 Figure 6-22. Histogram of the ratio of annual average NO<sub>2</sub> concentration (ppb) averaged 9 over a 3-year period from the contributing monitor with the maximum value 10 for each ecoregion to the average of weighted annual average NO<sub>2</sub> design 11 values (EAQM) over the same 3-year period.

### 6.2.2.4 PM<sub>2.5</sub> Results

2 Finally, similar analyses were also completed assessing the relationship between S, N, 3 and S+N deposition and air quality design value data for the current secondary PM<sub>2.5</sub> annual 4 standard.4 Figure 6-23 shows the relationship between upwind annual average PM<sub>2.5</sub> EAQM 5 data and S deposition levels over the usual five periods. The data points can be divided into two 6 groups. There are a minority of data pairs where S deposition is extremely low yet PM<sub>2.5</sub> EAQM 7 values are high. This is likely occuring in areas where the  $PM_{2.5}$  levels are driven by components 8 other than sulfate. Then there is a second set of data points where there is a positive association 9 between the upwind PM<sub>2.5</sub> EAQM and downwind S deposition. Overall, the correlation for the 10 paired data is 0.67, which falls between the range seen for the SO2 and NO2 EAQM data. Figure 11 6-24 describes the comparison between S deposition levels and the annual PM<sub>2.5</sub> DV from the 12 highest monitor in the ecoregions' sites of influence. The correlation between these two terms is 13 relatively low (r = 0.21). 14 However, there was very strong correlation between upwind PM2.5 EAQM and 15 downwind N deposition throughout the entire distribution (r = 0.98), as shown in Figure 6-25. 16 This strong correlation was diminished (r = 0.77) somewhat when moving from the weighted 17 EAQM to use of the maximum PM<sub>2.5</sub> DV from the highest monitor in the ecoregions' sites of influence (Figure 6-26). As shown in Figure 6-27, the ratios between the maximum PM<sub>2.5</sub> DV in 18 19 an ecoregion's sites of influence and the weighted EAQM value typically ranges from 1.11 to 20 1.66. Finally, Figures 6-28 and 6-29 illustrate the relationship between PM<sub>2.5</sub> design values and 21 total S+N deposition. The data suggest relatively strong correlation between PM<sub>2.5</sub> EAQM data 22 and total S+N deposition (r = 0.88), but less correlation with the maximum DV (r = 0.50).

<sup>&</sup>lt;sup>4</sup> Given the cumulative nature of N and S deposition, it was expected that an air concentration metric with a longer averaging time would be a more appropriate potential indicator of downwind deposition, thus the EPA restricted the PM<sub>2.5</sub> analysis to the annual standard and did not include analyses for the 24-hour standard.





Figure 6-23. Scatterplot of estimated 3-year average S deposition (ecoregion median) and
 the weighted annual average PM<sub>2.5</sub> design values from contributing upwind
 areas for that ecoregion (EAQM) also averaged over 3 years.





Figure 6-24. Scatterplot of estimated 3-year average S deposition (ecoregion median) and
 the average annual PM<sub>2.5</sub> design value over that 3-year period from the
 contributing monitor with the maximum value for each ecoregion.







Figure 6-25. Estimated 3-year average N deposition (ecoregion median) and average of
 weighted annual average PM<sub>2.5</sub> concentrations in 3-year period (EAQM) for
 that ecoregion.





Figure 6-26. Estimated 3-year average N deposition (ecoregion median) and annual
 average PM<sub>2.5</sub> concentration in 3-year period from maximum contributing
 monitor for that ecoregion.



Figure 6-27. Histogram of the ratio of average annual average PM<sub>2.5</sub> concentration (μg/m<sup>3</sup>) in 3-year period from maximum contributing monitor for that ecoregion to the average of weighted annual average PM<sub>2.5</sub> concentrations (EAQM) in 3-year period (median = 1.3).



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Figure 6-28. Estimated 3-year average S+N deposition (ecoregion median) and average of
 weighted annual average PM<sub>2.5</sub> concentrations in 3-year period (EAQM) for
 that ecoregion.



Figure 6-29. Estimated 3-year average S+N deposition (ecoregion median) and average annual average PM<sub>2.5</sub> concentration in 3-year period from maximum contributing monitor for that ecoregion.

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### 6.2.2.5 Conclusions

7 For SO<sub>2</sub>, we examined both the  $2^{nd}$  highest 3-hour maximum and an annual average 8 metric. The results for the EAQM suggest that both metrics are correlated with S deposition, 9 with the stronger association being for the annual average metric. There is lower correlation 10 between the design values from the highest monitor within the ecoregion sites of influence for 11 both the 2<sup>nd</sup> highest 3-hour maximum and an annual average SO<sub>2</sub> metrics. As shown by the ratio 12 information, this is likely due to the large concentration gradients seen across the SO<sub>2</sub> monitors 13 in the U.S. (for example, see Figure 2-23), with the maximum contributing monitor between 14 generally 3 to 4 times higher than the EAQM. These figures also show that in the most recent 15 assessed time period of 2018-2020, the median S deposition in the Ecoregion III areas was below 16 5 kg/ha-yr when the annual average  $SO_2$  concentration, averaged over three years, at contributing 17 monitors was less than 22 ppb and the majority of monitors were below 10 ppb. Additionally, the 18 SO<sub>2</sub> figures indicate that there can be high measured SO<sub>2</sub> concentrations associated with low S 19 deposition (i.e., < 5 kg S/ha-yr) and that there is generally more scatter in the data at lower 20 deposition values. Both of these observations could be due to uncertainties in the TDEP

calculations, uncertainties in our assessment methodology and/or a lack of correlation between
 some SO<sub>2</sub> monitor measurements and S deposition.

- 3 For NO<sub>2</sub>, the correlations between the measured annual NO<sub>2</sub> concentrations and N 4 deposition are not as strong as they are between metrics for SO<sub>2</sub> concentrations and S deposition. 5 This could be partially due to the fact that oxidized nitrogen only contributes to part of the total 6 N deposition estimate, and as discussed in section 2, the contribution of reduced nitrogen to total 7 N deposition has grown over the last few decades (e.g., Li et al., 2016). The figures also show 8 slightly less variability between the EAQM and maximum monitor concentrations for NO<sub>2</sub> 9 (when compared to  $SO_2$ ), with the  $NO_2$  maximum monitored values being typically about twice 10 as high as the calculated EAQM. This result suggests less variability and smaller gradients in 11 measured NO<sub>2</sub> concentrations across the U.S. when compared to SO<sub>2</sub>. In the most recent time 12 period (2018-2020), median N deposition was generally maintained at 12 kg/ha-yr in Ecoregion 13 III areas while NO<sub>2</sub> annual average, averaged over 3-years, monitored values were 30 ppb or 14 less. 15 For PM<sub>2.5</sub>, the assessment looks at correlations with S deposition, N deposition and S + N16 deposition. The results show a clear and remarkably strong correlation (r=0.98) between 17 measurements of annual average  $PM_{2.5}$  and estimates of N deposition. This could be due to 18 measurements at PM<sub>2.5</sub> monitors including both oxidized and reduced forms of N (i.e., NO<sub>3</sub> and 19 NH4<sup>+</sup>), which contribute together to total N deposition. While not as strong, there is a correlation 20 between measurements of annual average PM2.5 and estimates of S deposition. However, the 21 results include data where the measured  $PM_{2.5}$  mass is high when S deposition is low (i.e., < 2 kg) 22 S/ha-yr). This is similar to data seen in the figures assessing S deposition and SO<sub>2</sub> air quality 23 metrics. However, this could also be due to PM<sub>2.5</sub> mass at these contributing monitors having a 24 large fraction of non-S-containing compounds, such as NO<sub>3</sub><sup>-</sup>, NH<sub>4</sub><sup>+</sup> and/or organic carbon (OC). 25 In looking at the relationship between measurements of annual average PM<sub>2.5</sub> and estimates of 26 S+N deposition<sup>5</sup>, the results show a good correlation (r=0.88). For measurements of annual 27 average PM<sub>2.5</sub> there is less difference between the EAQM metric and the maximum monitor 28 concentrations for annual average PM<sub>2.5</sub>. In the most recent time period (2018-2020), PM<sub>2.5</sub> 29 annual average, averaged over 3-years, contributing monitored values were less than 18  $\mu$ g/m<sup>3</sup> 30 and mostly less than 15  $\mu$ g/m<sup>3</sup>, corresponding to N and S deposition of approximately 6-12 kg
- 31 N/ha-yr and <5 kg S/ha-yr, respectively.

<sup>&</sup>lt;sup>5</sup> Total deposition is converted to units of milli-equivalent using the following equation: S+N deposition = (6.25\*S deposition) + (7.14\*N deposition).

### 1 6.3 AIR QUALITY METRICS FOR CONSIDERATION

2 Based on the information above, this section discusses how well various air quality 3 metrics relate to S and N deposition. Section 6.2.1 examines this relationship in important 4 ecological areas of the country, with a focus on a subset of Class 1 areas. Generally, this section 5 looks at co-located information and includes data from monitors and models. Section 6.2.2 then 6 examines, for SO<sub>2</sub>, NO<sub>2</sub> and PM<sub>2.5</sub> design value or design value-like metrics, the relationship 7 between measured upwind air quality concentrations and eventual downwind S and N 8 deposition. This analysis is particularly relevant given that the current secondary standards are 9 judged using design value metrics based on measurements at the current  $SO_2$ ,  $NO_2$  and  $PM_{2.5}$ 10 FRM and FEM monitors. Most of these monitors are in the areas of higher pollutant 11 concentrations, and many are sited near  $SO_x$  and  $NO_x$  emissions sources. For example, as 12 discussed in section 2.3, many SO<sub>2</sub> monitors are sited near large point sources of SO<sub>2</sub> (e.g., 13 electric generating units) and for NO<sub>2</sub>, larger urban areas are required to site monitors to measure 14 NO<sub>2</sub> near larger roadways with a focus on mobile source emissions. Thus, this information can 15 help inform how changes in emissions relate to changes in deposition and how best to regulate 16 measured air quality concentrations through the NAAQS to maintain deposition at or below

17 certain levels.

### 18 **6.3.1 SO<sub>2</sub> Metrics**

As introduced in section 2, S tends to deposit as SO<sub>2</sub> close to sources of SO<sub>2</sub> emissions
but as SO<sub>4</sub> in areas further away, such as more rural areas of the country. In the western U.S.,
where S tends to be low, S may deposit more equally from SO<sub>2</sub> and SO<sub>4</sub><sup>2+</sup>.

22 Section 6.2.2 examines the current form and averaging time of the  $SO_2$  secondary NAAOS which is the 2<sup>nd</sup> highest 3-hour daily maximum for a year in the deposition to air quality 23 24 analyses. Additionally, given that the impacts examined in this review are associated with 25 deposition over some longer period of time (e.g., growing season, year, multi-year), section 6.2.2 26 also assesses an SO<sub>2</sub> air quality metric of an annual average. Additionally, noting the many 27 factors that can lead to variability in the deposition, including frequency of precipitation, and 28 micrometeorological factors relevant to the dry deposition velocity, the analyses focus on a 3-29 year average of all of the air quality and deposition metric and include multiple years of data to 30 better assess more typical relationships.

Based on the results of section 6.2.2, both the current standard form and averaging time,

32 as well as the annual average air quality metric, show a strong relationship with S deposition.

33 However, the annual average of SO<sub>2</sub>, averaged over 3-years, looks to have the strongest

34 correlation with S deposition averaged over 3-years. When further assessing these metrics, with a

35 focus on just the Ecoregion III areas used in the aquatic CL analyses (section 5.2) in Figures 6-30

- and 6-31, the same conclusions can be made. However, for these Ecoregion III areas there is less
- variability in the relationships, with a very strong correlation (r=0.94) between S deposition and
  - S Deposition, 3-yr average (kg S/ha-yr) 2001-2003 2006-2008 2010-2012 . . • 2014-2016 2018-2020 Weighted Annual Average SO2, average for 3-yr period
- annual average SO<sub>2</sub> averaged over 3-years.

Figure 6-30. For ecoregions included in the Aquatic CL Analysis, estimated 3-year average S deposition (ecoregion median) and weighted annual average SO<sub>2</sub> concentrations (EAQM) in 3-year period for that ecoregion (r=0.94).





Based on the information above, this section concludes that the quantitative analyses support using either one of the two air quality metrics assessed to control S deposition: (1) 2nd highest annual 3-hour daily maximum, averaged over 3-years or (2) annual average, averaged over three years. Between these two metrics, the SO<sub>2</sub> annual average, averaged over three years, would likely be the better choice given that the analyses show the metric to be more strongly related to S deposition.

13 When evaluating this information to assess a level at which one of these  $SO_2$  air quality 14 metrics might help maintain S deposition to an appropriate level, a few observations should be 15 considered. First, for SO<sub>2</sub>, the monitor concentrations can vary substantially across the U.S. This 16 is seen is the large ratio (i.e., 3-4) between the maximum contributing monitor concentration and 17 the EAQM. This large ratio means selecting a level based on the EAQM information alone will 18 lead to larger reductions than needed. Another observation is that there are a number of instances 19 where SO2 concentrations are high, but S deposition is low. This is generally seen at S 20 deposition values of less than 5 kg/ha-yr. There is also a substantial scatter at these lower 21 deposition values, calling into question the ability to select an SO<sub>2</sub> concentration level and metric 22 to maintain deposition below this 5 kg/ha-yr. However, it is worth noting that in the most recent

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assessed time period of 2018-2020, the median S deposition in the Ecoregion III areas was
 maintained below 5 kg/ha-yr when the annual average SO<sub>2</sub> concentration at contributing
 monitors, averaged over three years, was less than 22 ppb. The majority of monitors were below
 10 ppb.

5 6.3.2 NO<sub>2</sub> and PM<sub>2.5</sub> Metrics

6 For N, the results in section 6.2.1 suggest that oxidized N deposition in rural areas is 7 mostly from deposition of air concentrations of nitric acid and particulate nitrate, rather than 8 NO<sub>2</sub>. Additionally, the results suggest that in some areas inorganic nitrogen (e.g.,  $NH_4^+$ ) 9 contributes to the N deposition, with higher contributions in areas near emission sources of NH<sub>3</sub>. 10 Section 6.2.2 examines the current form and averaging time of the  $NO_2$  secondary 11 NAAQS which is the annual average NO<sub>2</sub> concentration. As in the assessments of the other 12 pollutants and air quality metrics, the analyses also focus on a 3-year average of NO<sub>2</sub> and N 13 deposition and include multiple years of data to better assess more typical relationships. For 14 NO<sub>2</sub>, the correlations between annual average NO<sub>2</sub> and N deposition were somewhat low (r=0.5815 for EAQM). In addition, the ratios between the maximum contributing monitor and the EAQM 16 show variability, though less than was seen for SO<sub>2</sub>, across the measured annual average 17 concentrations of NO<sub>2</sub> across the U.S., with a median ratio of 2. The correlation between annual 18 average PM<sub>2.5</sub> and N deposition was much stronger (r=0.98 for EAQM). This is likely due to 19 HNO<sub>3</sub>, NO<sub>3</sub> and NH<sub>4</sub><sup>+</sup> being the largest contributors to N deposition and being most closely 20 related to concentrations of  $PM_{2.5}$ . Additionally, the ratios between the maximum contributing 21 monitors and the EAQM are lower for  $PM_{2.5}$  (compared to  $SO_2$  and  $NO_2$ ) with ratios closer to 1 22 suggesting lower variability of annual average  $PM_{2.5}$  across the U.S. Given this information and 23 these relationships, the PM<sub>2.5</sub> annual average, averaged over three years, might be the better air 24 quality metric to control N deposition. Such a metric would also provide some control over S 25 deposition, as seen in the figures above. However, it is important to consider that this analysis 26 focuses on PM<sub>2.5</sub> monitors that contribute to the S and N deposition across the U.S. and that 27 these monitors (and other PM<sub>2.5</sub> monitors) also measure other non-S and N related pollutants as 28 part of the PM<sub>2.5</sub> total mass.

### 29 6.3.3 Key Uncertainties and Limitations

The linkage between air concentration and deposition can vary based on site-specific conditions, including the chemical form of nitrogen and sulfur, frequency of precipitation, and micrometeorological factors relevant to the dry deposition velocity. The analyses above attempt to provide insight into these relationships and variability for multiple measured air quality metrics. As with any assessment, there are uncertainties and limitations associated with the work,

1 most of which are discussed above in the context of each the analyses. In this section, we

2 summarize some of the overarching uncertainties and limitations.

3 In section 6.2.1, multiple forms of data were analyzed using co-located information in a 4 subset of Class I areas. While there are uncertainties in each of the different sets of modeled and 5 measured data analyzed, the fact that the assessment saw consistent results across these different 6 forms of data reduces the concern with these potential data-related issues. The biggest limitation 7 of the assessment in section 6.2.1 is the limited geographical coverage of the Class I areas that 8 were included. While these areas were selected from different parts of the country and were 9 chosen based on the availability of co-located air quality (i.e., IMPROVE, CASTNET) and 10 NADP/NTN monitors, most were located in the western U.S., where terrain, emissions and air 11 quality chemistry can look different from other parts of the country. This analysis may neglect or 12 underestimate the role of large ammonia emission sources in the Midwest and large nitrogen 13 oxide emission sources in the eastern U.S. Additionally, these selected Class I areas do not 14 include many of the locations that were quantitatively assessed in section 5 for potential aquatic 15 acidification effects, given that few are located in the eastern U.S. 16 In section 6.2.2, an analysis using the HYSPLIT model was included to assess the 17 linkage between TDEP estimates of N and S deposition and measured air quality concentrations of NO<sub>2</sub>, SO<sub>2</sub> and PM<sub>2.5</sub>. There are uncertainties in the HYSPLIT application itself, including the 18 19 use of one year of meteorological data to estimate multiple years of transport. Additionally, this 20 EPA analysis made subjective decisions as to what percentage of trajectory impacts warranted 21 inclusion in an ecosystem's sites of influence. It is unclear how much and in what way these 22 uncertainties and assumptions might impact the results. Although, increasing the geographic 23 scope of the sites of influence could lead to higher maximum values. There are also uncertainties 24 in the TDEP estimates, which are discussed in more detail in section 2.5. There is also 25 uncertainty as to whether only SO<sub>2</sub>, NO<sub>2</sub> and PM<sub>2.5</sub> concentrations at the monitor site influence 26 the designated receiving ecoregion deposition. An additional uncertainty that should also be

27 considered is the application of HYSPLIT to somewhat large area of the country (Ecoregion III

areas) which may have substantial spatial variability in deposition levels. In the analysis, a

29 median deposition level for each Ecoregion III area was used in considering the relationship

30 between deposition and air quality. To assess how these median data compare to those used in

31 the quantitative analysis (section 5.2), the median TDEP S deposition estimate for each

32 Ecoregion III area was compared to the median TDEP deposition estimate for each of the water

body locations used in each Ecoregion III area in the aquatic critical load analysis in section 5.

34 The comparison finds that the Ecoregion median can range from 31% higher to 22% lower than

35 the S deposition used in the aquatic analysis (and with a maximum difference of less than 3

36 kg/ha-yr) but, on average, is typically less than 7% different (see Appendix 6A, Table 6A-4).

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### 7 REVIEW OF THE STANDARDS

In considering what the currently available evidence and exposure/risk information
indicate with regard to the current secondary SO<sub>2</sub>, NO<sub>2</sub> and PM standards, the initial overarching
question we address is:

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## • Does the currently available scientific evidence and air quality and exposure analyses support or call into question the adequacy of the protection afforded by the current secondary standards?

8 To assist us in interpreting the currently available scientific evidence and quantitative 9 information, including results of recent and past quantitative analyses to address this question, 10 we have focused on a series of more specific questions. In considering the scientific and 11 technical information, we consider both the information previously available and information 12 newly available in this review which has been critically analyzed and characterized in the current 13 ISA, the 2008 ISA for the oxides of N and S, the 2009 PM ISA and prior AQCDs for all three 14 criteria pollutants. In so doing, an important consideration is whether the information newly 15 available in this review alters the EPA's overall conclusions from the last reviews regarding 16 ecological effects associated with oxides of N and S and PM in ambient air. We also consider the 17 currently available quantitative information regarding environmental exposures, characterized by 18 the pertinent metric, likely to be associated with the air quality metric representing the current 19 standards. Additionally, we consider the significance of these exposures with regard to the 20 potential for ecological effects, their potential severity and any associated public welfare 21 implications. 22 Within this chapter, the evidence and exposure-based questions regarding policy-relevant 23 aspects of the currently available information regarding effects, public welfare implications, the 24 current standards and as appropriate, consideration of potential alternatives are discussed in 25 sections 7.1 and 7.2. Section 7.1 addresses the questions in the context of direct effects of the

26 pollutants in ambient air and, in similar fashion, section 7.2 addresses policy-relevant questions

27 in the context of deposition related effects. Preliminary conclusions derived from the evaluations

- 28 presented in this draft PA are described in section 7.3. Section 7.4 identifies key uncertainties
- and areas for future research.

### 30 7.1 EVIDENCE AND EXPOSURE/RISK BASED CONSIDERATIONS 31 FOR DIRECT EFFECTS OF THE POLLUTANTS IN AMBIENT AIR

In considering the currently available evidence and quantitative information pertaining to
 direct effects of oxides of N and S and PM in ambient air, including what this information

indicates regarding effects, and associated public welfare implications, that might be expected to
 occur under air quality meeting the existing standards, we address the following questions.

- To what extent has the newly available information altered our scientific
   understanding of the direct welfare effects of oxides of S and N and PM in ambient
   air?
- To what extent does the currently available information indicate the potential for
   exposures associated with ecological effects under air quality meeting the existing
   standards? If so, might such effects be of sufficient magnitude, severity, extent
   and/or frequency such that they might reasonably be judged to be adverse to public
   welfare?
- 11 12

• To what extent have important uncertainties identified in past reviews been reduced and/or have new uncertainties emerged?

Framed by these questions, we consider the evidence and quantitative for the three criteriapollutants in the subsections below.

### 15 7.1.1 Direct Effects of SOx in Ambient Air

16 As summarized in section 4.1 above, very little of the currently available information 17 regarding the direct effects of SOx in ambient air is newly available in this review. Among the 18 SO<sub>X</sub>, which include SO, SO<sub>2</sub>, SO<sub>3</sub>, and S<sub>2</sub>O, only SO<sub>2</sub> is present in the lower troposphere at 19 concentrations relevant for environmental considerations (ISA, Appendix 2, section 2.1). Sulfate 20 is the prominent S oxide present in the particulate phase. The available evidence, largely 21 comprised of studies focused on SO<sub>2</sub>, documents its effects on vegetation, including foliar injury, 22 depressed photosynthesis and reduced growth or yield (ISA, Appendix 3, section 3.2). 23 In general, effects on plants occur at SO<sub>2</sub> exposures higher than a 3-hour average

concentration of 0.5 ppm. The evidence derives from a combination of laboratory studies and
observational studies. A recent laboratory study reports some transient effects on lichen

26 photosynthesis for short exposures, with more long-lasting effects only observed for exposures

of nearly 1 ppm SO<sub>2</sub>, as summarized in section 5.1.1 above. With regard to the sensitive effect of

28 foliar injury, the current ISA states there to be "no clear evidence of acute foliar injury below the

29 level of the current standard" (ISA, p. IS-37). Further, the "limited new research since 2008 adds

30 more evidence that SO<sub>2</sub> can have acute negative effects on vegetation but does not change

conclusions from the 2008 ISA regarding ... the SO<sub>2</sub> levels producing these effects" (ISA, p. IS37).

Uncertainties associated with the current information are generally similar to those of past reviews. In large part they relate to limitations of experimental studies in reflecting the natural environment and limitations of observational studies in untangling effects of SO<sub>2</sub> from those related to other pollutants that may have influenced the analyzed effects. Regardless of these uncertainties, we note that the evidence from either type of study indicates exposures
 associated with effects to generally be associated with air concentrations and durations which

3 would not be expected to occur when the current standard is met.

### 4 7.1.2 Direct Effects of N Oxides in Ambient Air

5 The currently available information on direct effects of N oxides in ambient air is 6 comprised predominantly of studies of NO<sub>2</sub> and HNO<sub>3</sub>, and also of PAN, with regard to effects 7 on plants and lichens (as summarized in section 4.1 above). The very few studies newly available 8 in this review do not alter our prior understanding of effects of these N oxides, include visible 9 foliar injury and effects on photosynthesis and growth at exposures considered high relative to 10 current levels in ambient air (ISA, section 3.3). Thus, as in the last review, the ISA again 11 concludes that the body of evidence is sufficient to infer a causal relationship between gas-phase 12 NO, NO<sub>2</sub>, and PAN and injury to vegetation" (ISA, section IS.4.2). Previously available 13 evidence for HNO<sub>3</sub> included experimental studies of leaf cuticle damage in tree seedlings, a 14 finding confirmed in a more recent study, as well as effects on lichens, as summarized in section 15 5.1.2 above. Effects of HNO<sub>3</sub> may be related to vapor exposures or deposition given its very high 16 deposition velocity (ISA, Appendix 3, section 3.4). The evidence includes studies of effects 17 related to historic conditions in the Los Angeles basin. A more recent 2008 reassessment of an 18 area in the Los Angeles basin in which there was a significant decline in species in the late 1970s 19 found that lichen communities have not recovered from the damage evident in the 1970s (ISA, 20 Appendix 3, section 3.4). The newer studies continue to support the findings of the 2008 ISA, 21 such that as in the last review, the ISA again concludes "the body of evidence is sufficient to 22 infer a causal relationship between gas-phase HNO<sub>3</sub> and changes to vegetation" (ISA, section 23 4.3).

24 With regard to the exposure concentrations, we note that for NO<sub>2</sub> "[w]ith few exceptions,

25 visible injury has not been reported at concentrations below 0.20 ppm, and these exceptions

26 occurred when the cumulative duration of exposures extended to 100 hours or longer" (ISA,

27 Appendix 3, p. 3-8). Effects on plant photosynthesis and growth have resulted from multiday

28 exposures of six or more hours per day to NO<sub>2</sub> concentrations above 0.1 ppm, with a newly

29 available study documenting effects at exposures of 4 ppm NO<sub>2</sub> which are "consistent with past

30 studies of plants with relatively high NO<sub>2</sub> exposure" (ISA, Appendix 3, pp. 3-12). Regarding

31 PAN, there is "little evidence in recent years to suggest that PAN poses a significant risk to

32 vegetation in the U.S." (ISA, Appendix 3, p. 3-13). The recently available information for HNO<sub>3</sub>

33 includes effects on tree foliage under controlled 12-hour exposures to 50 ppb HNO<sub>3</sub>

34 (approximately 75 µg/m<sup>3</sup>). Foliar damage was also reported in longer, 32- or 33-day exposures in

35 which peak HNO<sub>3</sub> concentrations for the "moderate" treatment (30-60  $\mu$ g/m<sup>3</sup>) encompassed the

1 range reported in summers during the 1980s in the Los Angeles Basin, as described in section 2 5.1.2 above (ISA, Appendix 3, section 3.4;). During that period,  $NO_2$  concentrations in the Basin 3 range up to 0.058 ppm, exceeding the secondary standard (U.S. EPA, 1987). Effects on lichen 4 photosynthesis have been reported from 6.5-hour daily varying exposures with peaks near 50 ppb 5  $(\sim 75 \ \mu g/m^3)$  that extend beyond 18 days (ISA, Appendix 6, section 6.2.3.3; Riddell et al., 2012). 6 In considering the potential for concentrations of N oxides associated with welfare effects 7 to occur under air quality conditions meeting the current NO<sub>2</sub> standard, we consider the air 8 quality information summarized in section 2.4.1 above. In so doing, we note that air quality at all 9 sites in the U.S. have met the existing secondary standard since around 1991 (Figure 2-21). During the period 1983 to 1991, the 99<sup>th</sup> percentile of annual mean NO<sub>2</sub> concentrations at sites 10 11 nationwide was near the level of the standard (Figure 2-21). Further, hourly NO<sub>2</sub> concentrations 12 during this time indicate little likelihood of an occurrence of a 6-hour concentration of magnitude 13 for which plant growth effects were reported from experimental studies (as described in section 5.1.2), as the 98<sup>th</sup> percentile of 1-hour concentrations rarely exceeded 0.2 ppm, as shown in 14 15 Figure 2-20. With regard to the potential for HNO<sub>3</sub> concentrations occurring in conditions that 16 meet the current  $NO_2$  secondary standards to pose risk of effects, we consider the larger evidence 17 base in that regard. As assessed in the ISA, the evidence indicates NO<sub>2</sub>, and particularly, HNO<sub>3</sub>, 18 as "the main agent of decline of lichen in the Los Angeles basin" (ISA, Appendix 3, p. 3-15), 19 thus indicating a role for the elevated concentrations of N oxides documented during the 1970s 20 to 1990s (and likely also occurring earlier). Based on studies extending back to the 1980s, HNO3 21 has been suspected to have had an important role in these declines, as summarized in section 22 5.1.2 above. During that time period the Los Angeles metropolitan area experienced NO<sub>2</sub> 23 concentrations well in excess of the NO<sub>2</sub> secondary standard. For example, annual average NO<sub>2</sub> 24 concentrations in Los Angeles ranged up to 0.078 ppm in 1979 and remained above the standard 25 level of .053 ppm into the early 1990s (Appendix 5B, section 5B.4.1). A resampling in the Los 26 Angeles Basin in 2008 reported that the impacts documented on lichen communities in the 1970s 27 remained at that time (ISA, Appendix 3, section 3.4). The extent to which this relates to lag in 28 recovery or concentrations of various air pollutants is unknown. Thus, the currently available 29 information is limited with regard to the extent to which it informs conclusions as to the potential 30 for exposures associated with ecological effects under air quality meeting the existing NO<sub>2</sub> 31 secondary. More recent studies extending into more recent periods indicate variation in eutrophic 32 lichen abundance to be associated with variation in metrics representing N deposition (ISA, 33 Appendix 6, section 6.2.3.3). The extent to which these associations are influenced by residual 34 impacts of the historic air quality is unclear.

35 While new uncertainties have not emerged, uncertainties remain in our interpretation of 36 the evidence. These include those related to limitations of the various study types. For example,

1 the various types of studies in the evidence for welfare effects of the different N oxides vary with 2 regard to their limitations, and associated uncertainties. Field studies are limited with regard to 3 identification of threshold exposures for the reported effects and uncertainties associated with 4 controlled experiments include whether the conditions under which the observed effects occur 5 would be expected in the field. A key uncertainty affecting interpretation of studies of historic 6 conditions in the LA Basin relates to the extent to which other air pollutants or local conditions 7 may have contributed to the observations. With regard to the risk posed by N oxides, and 8 particularly  $HNO_3$ , the evidence, as summarized in sections 5.1.2 and 5.4.3.2 above indicates the 9 potential for effects of air quality occurring during periods when the current secondary standard 10 was not met, which, depending on policy judgments, may be concluded to have public welfare 11 implications. The evidence is limited, however, with regard to support for conclusions related to 12 conditions meeting the current standard.

### 13 7.1.3 Particulate Matter

As summarized in section 5.1.3 above, the evidence for ecological effects of PM is consistent with that available in the last review. The ISA causal determinations with regard to ecological effects of PM in the last PM review and in this review focused on deposition-related effects, rather than direct effects of PM in ambient air. In this review, as in the last one, the ecological effects evidence were found to be sufficient to conclude there is likely to exist a causal relationship between deposition of PM and a variety of effects on individual organisms and ecosystems (ISA, Appendix 15; 2012 PM ISA, section 9.4).

With regard to direct effects of PM in ambient air, the information on ambient air concentrations is well in excess of the existing secondary standards. While some uncertainties remain, new uncertainties have not emerged since the last review. In summary, little information is available on welfare effects of PM in exposure conditions likely to meet the current standards, and that which is does not indicate effects to occur under those conditions.

### 26 7.2 EVIDENCE AND EXPOSURE/RISK-BASED CONSIDERATIONS 27 FOR DEPOSITION-RELATED EFFECTS

In this section, we consider the evidence and quantitative exposure/risk information related to deposition-related ecosystem effects of oxides of S and N and PM in ambient air. We do this in the larger context of evaluating the protection from such effects provided by the existing standards and potential alternative standards. The potential for the three criteria pollutants to all contribute to particular ecosystem effects while also having a potential for independent effects poses challenges to the organization of the discussion. While recognizing there are multiple organizations that could be applied, we have adopted one that focuses first on

1 consideration of S deposition (section 7.2.1) and then N deposition (section 7.2.2). Further,

2 within each of these sections, we first consider the evidence regarding deposition effects and the

3 evidence to support analysis of deposition levels associated with effects of potential public

4 welfare significance, and then the consideration of deposition levels that may be appropriate to

5 target for consideration of the public welfare protection appropriately afforded by the secondary

- 6 standards.
- 7

### 7.2.1 S Deposition and Oxides of S

8 To inform conclusions in this review related to the  $SO_x$  secondary standards, we consider 9 a series of questions below that are intended to facilitate the evaluation of the linkages between S 10 oxides in ambient air with S deposition and its associated welfare effects. In considering these 11 questions, we draw on the available welfare effects evidence described in the current ISA, the 12 2008 NO<sub>x</sub>/SO<sub>x</sub> ISA, the 2009 PM ISA, and past AQCDs, and summarized in chapter 4 above. 13 We do this in combination with the available information from quantitative analyses (and 14 summarized in Chapters 5 and 6 above), both analyses recently developed and those available 15 from the 2009 REA and considering the information now available.

### 16 7.2.1.1 Welfare Effects Evidence of Deposition-Related Effects

17 The long-standing evidence documents the array of aquatic and terrestrial effects of S and 18 acidic deposition. This evidence, extending back many decades, has accrued in part through 19 study of ecosystem acidification that has resulted from historic acid deposition. As discussed in 20 prior chapters, both S and N compounds have contributed, with relative contributions varying 21 with both emissions, air concentrations and atmospheric chemistry, among other factors. The 22 ecosystem effects, documented comprehensively in waterbodies of the Adirondack and 23 Appalachian Mountains, and forests of the northeast, have ranged from the organism to 24 ecosystem-level scale. The focus in this chapter is on considering quantitative aspects of the 25 relationships between deposition and ecosystem effects that can inform decisions on standards 26 that provide the appropriate control on deposition for the desired level of protection from adverse 27 environmental effects. 28 As recognized in Chapter 5, and the associated appendices, the availability of quantitative

29 information for relating atmospheric deposition to specific welfare effects varies across the

30 categories of effects. We consider here the extent to which such information is available that

- 31 might provide support to characterization of the potential for effects, and of the protection that
- 32 might be afforded for such effects, under different air quality conditions. We do this in the

33 context of the following question.

• To what extent does the currently available evidence base provide established quantitative approaches for characterizing ecosystem responses to S deposition that can inform judgments on the risk or likelihood of occurrence of ecosystem effects under differing conditions of SO<sub>x</sub> air quality?

5 The currently available information provides modeling approaches for quantitatively 6 analyzing linkages between S deposition, geochemical processes in soils and waterbodies and 7 indicators of aquatic and terrestrial ecosystem acidification risk. Aspects of the modeling 8 approaches that quantify processes that are the major determinants of the indicators have been 9 expanded and improved since the last review. Further, use of such modeling approaches for 10 characterizing potential risk of aquatic and terrestrial acidification is well established. Modeling 11 approaches vary in their complexity, precision, and limitations. Similarly, the evidence base 12 supporting risk characterization using the different acidification indicators also varies, with 13 associated uncertainties.

14 As recognized in Chapter 5 above, we have greatest confidence in the approach and tools 15 applied in the assessment of aquatic acidification. Although the approaches and tools for 16 assessing aquatic acidification are more generally utilized for S and N deposition in combination, 17 the approach taken in the analysis of aquatic ecosystem acidification summarized in section 5.2 18 above was to focus on S deposition. This decision was based on analyses indicating the relatively 19 greater role of S deposition under the more recent air quality conditions (as summarized in 20 section 5.2.1.4 above). The aquatic acidification assessment utilizes site-specific water quality 21 modeling that relates atmospheric deposition to ANC in a CL-based approach, as summarized in 22 section 5.2 above and described in more detail in Appendix 5A. The site-specific modeling 23 applications and associated estimates of CLs for different ANC targets are publicly available in 24 the NCLD. The modeling applications generally utilize mass balance and dynamic modeling 25 tools for watershed processes (e.g., fluxes that affect watershed concentrations of anions and 26 cations). In summary, the aquatic acidification assessment has utilized well-established site-27 specific water quality modeling applications with a widely recognized indicator of aquatic 28 acidification.

29 Quantitative tools are also available for the assessment of terrestrial acidification related 30 to S deposition (using BC:Al ratio as an indicator), as they were in the last review (section 31 5.4.2.1; 2009 REA, section 4.3). Recently available studies have addressed a particular area of 32 uncertainty identified for this approach in the last review (related to model inputs for base cation 33 weathering). While updated analyses have not been performed in this review, the findings from 34 the 2009 analyses, have been considered in the context of more recently available evidence 35 (section 5.4.2.1; 2009 REA, section 4.3). Quantitative tools and approaches are not as developed

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1 for other deposition-related effects associated with SO<sub>x</sub> in ambient air, such as mercury

2 methylation and sulfide toxicity (summarized in sections 4.2.3.1 and 4.2.3.2 above).

3 In summary, as in the last review, we find the quantitative approaches and tools for 4 assessment of aquatic acidification (including that attributable to S deposition) to be the most 5 advanced. While recognizing the uncertainties associated with results of analyses utilizing these 6 tools, as described in section 5.2 above, we recognize these results to be informative to our 7 purposes in identifying S deposition benchmarks associated with potential for aquatic 8 acidification effects of concern. As described in section 3.3.2 above, this assessment of 9 quantitative linkages between S deposition and potential for aquatic acidification is one 10 component of the approach implemented in this PA for informing judgments on the likelihood of 11 occurrence of such effects under differing air quality conditions.

12 7.2.1.2 General Approach for Considering Public Welfare Protection

### • To what extent does the available evidence support the use of waterbody ANC for purposes of judging a potential for ecosystem acidification effects?

15 As described in section 5.2.1 above, ANC is an indicator of susceptibility or risk of 16 acidification-related effects in waterbodies. Accordingly, the evidence generally indicates that the higher the ANC, the lower the potential for acidification and related waterbody effects, and 17 18 the lower the ANC, the higher the potential. The support for this relationship is strongest in 19 aquatic systems low in organic material, and the evidence comes predominantly from impacted 20 waterbodies in the eastern U.S. and Canada. In waterbodies with high dissolved organic material 21 (e.g., dissolved organic carbon), however, while the organic acid anions contribute to reduced 22 pH, these anions create complexes with the dissolved Al, protecting resident biota against Al 23 toxicity (ISA, Appendix 8, section 8.3.6.2). Accordingly, biota in such systems tolerate lower 24 ANC values than biota in waterbodies with low DOC. Thus, while the evidence supports the use 25 of ANC as an acidification indicator, the relationship with risk to biota differs depending on the 26 presence of naturally occurring organic acids. Further, such natural acidity affects the 27 responsiveness of ANC to acid deposition in these areas. As noted in section 5.2 above, the 28 ecoregions in which ANC is less well supported as an indicator for acidic deposition-related 29 effects due to the prevalence of waterbodies with high dissolved organic material include the 30 Middle Atlantic Coastal Plain (ecoregion 8.5.1), Southern Coastal Plains (ecoregion 8.5.3), and 31 Atlantic Coastal Pine Barrens (ecoregion 8.5.4). The evidence does, however, support the use of 32 waterbody ANC for purposes of judging a potential for ecosystem acidification effects (section 33 5.2.2.2). 34 As summarized in sections 4.2.1.1.2 and 5.2.1 above, there is longstanding evidence of an

35 array of significant impacts on aquatic biota and species richness reported in surface waters with

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1 ANC values below zero, and in waters with ANC values below 20  $\mu$ eq/L. This evidence derives

- 2 primarily from lakes and streams of the Adirondack Mountains and areas along the Appalachian
- 3 Mountains. The evidence base additionally indicates increased risk to resident biota of ANC
- 4 levels between 20 and 50  $\mu$ eq/L, as summarized in section 5.2.1 above. As recognized in the last
- 5 review, in addition to providing protection during base flow situations, ANC is a water quality
- 6 characteristic that affords protection against the likelihood of decreased pH from episodic events
- 7 in impacted watersheds. For example, waterbodies with ANC below 20 µeq/L have been
- 8 generally associated with high probability of low pH events, that have potential for death or loss
- 9 of fitness of sensitive biota (2008 ISA, section 5.2.2.1). In general, the higher the ANC level
- 10 above zero, the lower the risk presented by episodic acidity. In summary, the available evidence
- 11 provides strong support for the consideration of ANC for purposes of making judgments
- 12 regarding risk to aquatic biota in acid deposition impacted streams, and for consideration of the
- 13 set of targets analyzed in the quantitative aquatic acidification assessment: 20, 30, and 50  $\mu$ eq/L
- 14 (section 5.2 above).

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## • To what extent might waterbodies in sensitive ecoregions experiencing S deposition across the range of recent time periods be expected to achieve ANC levels of interest? What are associated uncertainties in these estimates?

18 In considering this question, we focus on the results of the quantitative aquatic 19 acidification assessment at three scales: national-scale, ecoregion-scale and focused case study-20 scale, as described in section 5.2 above. An array of approaches are employed across the three 21 scales, all of which make use of water quality modeling-based CLs derived for three different 22 ANC targets. We give particular focus to the ecoregion and case-study analyses, which utilize 23 the waterbody-specific comparisons of estimated deposition and waterbody CLs to provide 24 ecoregion wide and cross-ecoregion summaries of estimated waterbody responses to ecoregion 25 estimates of deposition. In so doing, we have considered the extent to which waterbodies in each 26 ecoregion analyzed were estimated to achieve ANC levels at or above each of the three targets. 27 In this way we recognize the variation in ANC response across waterbodies in an ecoregion that 28 may be reasonable to expect with different patterns of S deposition. We also recognize that S 29 deposition that may be controlled by one or more NAAQS will vary across the U.S. such that 30 implementation of any new concentration based standard would be associated with a distribution 31 of different deposition levels across the U.S. 32 The national-scale analysis involved the 13,824 waterbodies for which a CL based on 33 ANC target was available. Unlike the case for the 2000-02 period analyzed in the last review,

34 few waterbodies are estimated to be receiving deposition in excess of their critical loads for

35 relevant ANC targets under recent deposition levels. More specifically, under deposition

1 estimated for the most recent time period (2018-2020), generally below 4 kg/ha-yr, only 4% of 2 waterbodies nationally were estimated to exceed CLs for an ANC of 50  $\mu$ eq/L (Table 5-1). 3 The ecoregion analyses provided a dataset of five ecosystem deposition estimates (for the 4 five time periods from 2001-03 to 2018-20) for each of 18 eastern ecoregions that has been 5 summarized in terms of percentage of waterbodies estimated to achieve ANC at or above the 6 three ANC targets. We focused primarily on the results for the deposition bins representing half 7 or more of the full dataset (those for 5 kg/h-yr and for higher levels). Across the dataset of 90 8 combinations of eastern ecoregions and deposition time periods, with CL exceedances organized 9 by deposition bins, with the highest being S deposition at or below 18 kg/ha-yr, at least 90% of 10 waterbodies per ecoregion were estimated to achieve ANC at or above 20 µeq/L in 73% of the 11 ecoregion-time period combinations, and at or above 50  $\mu$ eq/L in 60% of the combinations 12 (Table 5-4). This summary contrasts with that for the 76 combinations for S deposition at or 13 below 11 kg/ha-yr, for which at least 90% of waterbodies per ecoregion were estimated to 14 achieve ANC at or above 20 µeq/L in 83% of the combinations, and with that for the 69 15 combinations for S deposition at or below 9 kg/ha-yr, for which at least 90% of waterbodies per 16 ecoregion were estimated to achieve an ANC at or above 20 µeq/L in 87% of the combinations, 17 and at or above 50  $\mu$ eq/L in 72% of the combinations (Table 5-4). Although fewer ecoregion-18 time period combinations are associated with still lower S deposition estimates, contributing to 19 increased uncertainty, we also note that for the 63 ecoregion-time periods for which S deposition 20 is estimated at or below 7 kg/ha-yr, at least 90% of waterbodies per ecoregion were estimated to 21 achieve an ANC at or above 20  $\mu$ eq/L in 92% of the combinations, and at or above 50  $\mu$ eq/L in 22 78% of the combinations (Table 5-4). Lastly, for the lowest bin comprised of at least half of the 23 full dataset, of the 51 ecoregion-time periods with S deposition estimates at or below 5 kg/ha-yr, 24 90% of waterbodies per ecoregion were estimated to achieve an ANC at or above 20  $\mu$ eq/L in 25 96% of the combinations, and at or above 50  $\mu$ eq/L in 82% of the combinations 26 The ecoregion analysis results summarized for the deposition estimate bins at or below 27 11 kg/ha-yr (and at/below lower values), indicate the likelihood of appreciable improvements in 28 waterbody buffering capacity compared to that estimated for the set of ecoregion-time periods 29 reflecting deposition estimates as high as 18 kg/ha-yr. This improvement includes an appreciably 30 increased number of waterbodies in more ecoregions achieving ANC at or above 20 µeq/L, at or 31 above 30  $\mu$ eq/L and also at or above 50  $\mu$ eq/L. Additionally, these estimates increase with bins 32 for lower deposition estimates, while also representing reductions in the size of the supporting 33 dataset.

In the case study analyses, CL estimates for ANC targets of 20, 30 and 50 µeq/L are
summarized for waterbodies in five sensitive areas, three areas in the eastern U.S. and two in the
western U.S. (Table 5-5). For the Shenandoah National Park, one of the Class I areas, and the

1 study area for which there are CLs available in the NCLD for all 4977 waterbody sites

- 2 (McDonnell et al., 2014), 70% of the area's waterbodies are estimated to be able to achieve an
- 3 ANC at or above 20 µeq/L with annual average S deposition of 9.4 kg/h-yr; the comparable
- 4 value for 90% of the waterbodies in 7.1 kg/h-yr. The S deposition values for the 70<sup>th</sup> and 90<sup>th</sup>
- 5 percentile of waterbody CLs for the other less well studied areas, for which there are fewer
- 6 waterbodies for which modeling has been performed to estimate CLs, were consistently lower.

7 And, as one example of variability in estimates, and associated uncertainties, we observe that the

- 8 70<sup>th</sup> and 90<sup>th</sup> percentile waterbody CL estimates for an ANC target of 20  $\mu$ eq/L for the Sierra
- 9 Nevada study area, a much less well studied area than the eastern areas, are appreciably lower
- 10 than such estimates for all of the other case studies for any of the three ANC targets. Yet, we
- 11 note that the average of waterbody CLs for achieving ANC at or above each of the three targets
- 12 (20, 30 or 50  $\mu$ eq/L) within each of the five case studies were quite similar, ranging only from
- 13 9.4 to 12 kg/ha-yr.

14 In summary, the array of CL-based analyses provides a general sense of the ANC values 15 that waterbodies in sensitive regions across the continental U.S. may be able to achieve, 16 including for areas heavily impacted by a long history of acid deposition, such as waterbodies in 17 the Shenandoah National Park. In considering this information we also note the uncertainties 18 associated with such estimates, as in the last review. These include uncertainties associated with 19 the estimation of the ANC levels that individual waterbodies might be expected to achieve under 20 different rates of S deposition. This estimation is based on site-specific steady-state water quality 21 modeling, with associated limitations and uncertainties. For example, as recognized in sections 22 4.2.1.1.3 and 5.2.4 above, the data to support the site-specific model inputs for some areas are 23 more limited than others, with associated greater uncertainties. Further, there are additional 24 uncertainties associated with the estimates of S deposition for use in the analyses of CL exceedances, such as those for the national- and ecoregion-scale analyses. Consideration of such 25 26 uncertainties informs the weighing of the findings of the quantitative analyses. For example, in 27 light of the variation in uncertainty associated with the more to less well studied areas may 28 indicate the appropriateness of a greater emphasis on the former and/or less emphasis on 29 estimates for the upper end of the distribution.

# What does the quantitative information regarding S deposition and terrestrial acidification indicate regarding deposition levels of relatively greater and lesser concern as to the potential for acidification-related effects? What are associated uncertainties?

As recognized in Chapter 5, the quantitative tools for characterizing waterbody response
 to acidic deposition are well established and/or have been extensively applied in a greater variety
 of locations. Further, there is much greater availability of site-specific water quality

1 measurements than of soil quality measurements in sensitive areas across the U.S. The available

2 quantitative information related to terrestrial acidification summarized in Chapter 5 (and

3 presented in more detail on Appendix 5B) includes discussion of soil chemistry modeling

4 analyses (both those described in published studies and an analysis performed in the 2012 oxides

5 of N and S review), studies involving experimental additions of S compounds to defined forestry

6 plots, and observational studies of potential relationships between terrestrial biota assessments

7 and metrics for S deposition. We consider each here in consideration of the questions posed

8 above.

9 With regard to soil chemistry modeling, we note first the quantitative analyses performed 10 in the last review of soil acidification in areas of the northeastern U.S. in which two sensitive tree 11 species, sugar maple and red spruce, are widely distributed. These analyses yielded estimates of 12 acidic deposition CLs for several targets for a well-studied indicator of soil acidification, BC:Al 13 ratio (2009 REA, section 4.3). These estimates indicated a range of annual deposition rates under 14 which ratios at or above the intermediate target value of 1 were well above the CL estimates 15 associated with achieving various ANC targets in the aquatic acidification analyses discussed 16 above, and also above all of the ecoregion estimates (across the five time periods from 2001 17 through 2020) considered in the aquatic acidification analyses (Table 5-6). This is also the case 18 for the most conservative ratio value of 10. As concluded in the 2009 REA, an important source 19 of uncertainty in the simple mass balance model used in the analysis is the soil weathering 20 parameter (as is also the case in water quality modeling). We additionally note that published 21 studies since the 2009 REA, including one focused on areas of Pennsylvania, have utilized 22 different estimates for this parameter intended to reduce the associated uncertainty and have 23 reported somewhat higher CL estimates when the updated approach is used.

With regard to the information available from studies involving S additions to
experimental forested areas, we note, as an initial matter, the somewhat limited number of tree
species that have been included in such experiments. Although limited in number, the more
widely recognized sensitive species, from field observations, have been included in such studies.
We note that the available studies have not reported effects on the trees analyzed with additions
below 20 kg/ha-yr (in addition to the background atmospheric addition during the experiment).

30 The recently available quantitative information regarding S deposition and terrestrial 31 acidification also includes two observational studies that report associations of tree growth 32 and/or survival metrics with various air quality or S deposition metrics, providing support to 33 conclusions regarding the role of acidic S deposition on tree health in the U.S., most particularly 34 in regions of the eastern U.S. (section 5.4.2.3 and Appendix 5B, section 5B.3.2). The metrics 35 utilized in the two largest studies include site-specific estimates of average  $SO_4^{2-}$  deposition and 36 of average total S deposition over the interval between tree measurements, generally on the order

of 10 years (Dietze and Moorcroft, 2011; Horn et al., 2018). In the study that used SO<sub>4</sub><sup>2-</sup> as the 1

- 2 indicator of acidic S deposition, and for which the study area was the eastern half of the
- contiguous U.S., site-specific average  $SO_4^{2}$  deposition (1994-2005) ranged from a minimum of 3
- 4 4 kg/ha-yr to a maximum of 30 kg/ha-yr (Dietze and Moorcroft, 2011). Review of the study area
- 5 for this study and a map indicating geographic patterns of deposition during the period of the
- 6 deposition data indicate the lowest deposition areas to be farthest western, northeastern and
- 7 southeastern areas of the eastern U.S. (in which S deposition in the 2000-2002 period is
- 8 estimated to fall below 8 kg/ha-yr), and the highest deposition areas to be a large area extending
- 9 from New York through the Ohio River valley (Appendix 5B, Figures 5B-1 and 5B-11).
- 10 Deposition at the sites with species for which growth or survival was negatively associated with
- 11 S deposition in the second study ranged from a minimum below 5 kg/ha-yr to a site maximum
- 12 above 40 kg/ha-yr, with medians for these species generally ranged from around 5 to 12 kg/hr-yr
- 13 (Appendix 5B, section 5B.3.2.3; Horn et al., 2018).
- 14 As recognized in section 5.4.2 and Appendix 5B, the history of appreciable acidic
- 15 deposition in the eastern U.S. and the potential for the deposition patterns (e.g., locations of 16
- relatively greater versus relatively lesser deposition) to be somewhat similar may be an influence 17
- in the findings. This indicates an uncertainty with regard to the specific magnitude of deposition
- 18 that might be expected to elicit specific tree responses, such as those for which associations have 19
- been found. As recognized by the study by Dietze and Moorcroft (2011), which grouped species 20
- into plant functional groups, acidification impacts on tree mortality result from cumulative long-
- 21 term deposition and patterns reported by their study should be interpreted with that in mind.

#### 22 7.2.1.3 Relating Deposition-related Effects to Air Quality Metrics

23 In this review, we have explored how well various air quality metrics relate to S and N 24 deposition. The analyses examine, for design value or design value-like metrics, the relationship 25 between measured air quality concentrations and transported S and N deposition. This analysis is 26 particularly relevant given that the current secondary standards are judged using design value 27 metrics based on measurements at existing SO<sub>2</sub>, NO<sub>2</sub> and PM<sub>2.5</sub> FRM/FEM monitor locations. 28 Most of these monitors are located in areas of higher pollutant concentrations near emissions 29 sources. For example, many SO<sub>2</sub> monitors are sited near large point sources of SO<sub>2</sub> (e.g., electric 30 generating units). Accordingly, information from these monitoring sites can help inform how 31 changes in SO<sub>2</sub> emissions relate to changes in deposition and, correspondingly, what secondary 32 standard options might best regulate ambient air concentrations such that deposition in areas of 33 interest is maintained at or below certain levels. The details of these analyses are described in

- 34 Chapter 6 and Appendix 6A. In addressing the questions below, we consider the findings of
- 35 those analyses specific to S deposition associated with SOx and PM.

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## • What does the available information and air quality analyses indicate regarding relationships between air quality metrics related to the existing standards and S deposition? What are the uncertainties in relationships using such metrics?

There are many factors contributing to temporal and spatial variability in S deposition, including frequency of precipitation, and micrometeorological factors relevant to the dry deposition velocity. Based on the air quality information and analyses in Chapter 2 and 6, we find that S tends to deposit as SO<sub>2</sub> (in dry deposition) close to sources of SO<sub>2</sub> emissions and as SO<sub>4</sub> in areas further away, such as in the more rural areas of the country. In the western U.S., where S tends to be low, S may deposit more equally from SO<sub>2</sub> and SO<sub>4</sub>.

10 The analyses in Chapter 2 and 6 assess  $SO_2$  concentrations using a metric based on the current form and averaging time of the secondary SO<sub>2</sub> NAAQS, which is the 2<sup>nd</sup> highest 3-hour 11 12 daily maximum in a year, as well as an annual average SO<sub>2</sub> air quality metric. Additionally, in 13 light of the many factors contributing variability to S deposition, the analyses focus on a 3-year 14 average of all of the air quality and deposition metrics and include multiple years of data to 15 better assess more typical relationships. Specifically, the assessment includes data spanning 20 16 years, with a focus on the following set of 3-year periods: 2001-2003, 2006-2008, 2010-2012, 17 2014-2016 and 2018-2020.

18 The results suggest that a standard in the form of either metric analyzed (i.e., the 2nd 19 highest 3-hour maximum in a year, averaged over 3 years or an annual average, averaged over 3 20 years) might be expected to provide a level of control of S deposition across the U.S. 21 Additionally, of these two air quality metrics, the analyses suggest a potential for better control 22 with the annual average of SO<sub>2</sub> concentrations, averaged over 3 years, given that the analyses 23 show this metric to be more strongly related to S deposition. This potential for better control 24 notwithstanding, we take note of two additional considerations. First, monitor concentrations of 25 SO<sub>2</sub> can vary substantially across the U.S., in response to source emissions, complicating 26 consideration of how the maximum contributing monitor (as identified in the HYSPLIT analysis 27 described in section 6.2.2 above) relates to S deposition levels in downwind ecosystems. This 28 consideration is integral to identifying levels for a potential alternate standard that would avoid 29 over- (or under-) control. The other consideration is the finding of a number of instances in the 30 full dataset (spanning 20 years) of low S deposition associated with relatively higher SO<sub>2</sub> 31 concentrations. This generally involved S deposition values below 5 kg/ha-yr. At these lower 32 values, there is a substantial amount of scatter in the relationship between measured SO<sub>2</sub> 33 concentration and S deposition estimates, contributing increased uncertainty to the identification 34 of a levels for a SO<sub>2</sub> metric for a potential secondary standard that might be expected to maintain 35 deposition at or below 5 kg/ha-yr. This scatter could relate uncertainties in the TDEP estimates, 36 particularly given that many of these sites tend to be in the western U.S. For these lower

1 deposition values we find it appropriate to rely to a greater extent on air quality relationships

- 2 observed more recently. For example, for the most recent time period analyzed (i.e., 2018-2020),
- 3 the median S deposition in the Ecoregion III areas was maintained below 5 kg/ha-yr when the
- 4 annual average SO<sub>2</sub> concentration, averaged over three years, at contributing monitors was less
- 5 than 22 ppb and the majority of monitors were below 10 ppb.
- 6 The analyses for  $PM_{2.5}$  show a positive relationship between measurements of annual 7 average  $PM_{2.5}$  and estimates of S deposition. However, similar to the SO<sub>2</sub> air quality metrics, the 8 results also show that the measured  $PM_{2.5}$  mass can be high when S deposition is quite low (i.e.,
- 9 <2 kg S/ha-yr). This could be due to  $PM_{2.5}$  mass at these contributing monitors being dominated
- by non-S-containing compounds, such as NO<sub>3</sub>, NH<sub>4</sub> and/or organic carbon (OC). However, it is
   worth noting that in a recent time period (2018-2020), median Ecoregion III area S deposition
- worth noting that in a recent time period (2018-2020), median Ecoregion III area S deposition
  levels were at or below 5 kg S/ha-y when the PM<sub>2.5</sub> annual standard levels at contributing
- 12 reversivere at or below 5 kg 5/na-y when the 1 M<sub>2.5</sub> annual standard revers at contributing
- 13 monitors were generally less than 15  $\mu$ g/m<sup>3</sup> (i.e., the level of the current annual average,
- 14 secondary standard for PM<sub>2.5</sub>).

15 Lastly, as recognized in Chapter 6, we note multiple uncertainties with the analyses 16 approach that are important to interpretation of the results. It is unclear, however, how much and 17 in what way each of these uncertainties might impact the results. There are also uncertainties 18 inherent in the derivation of the TDEP estimates, which are discussed in more detail in Chapter 19 2. Another important uncertainty is associated with application of the HYSPLIT model to predict 20 transport trajectories between monitor locations and Ecoregion III areas, as well as the use of the 21 median TDEP deposition estimates across each Ecoregion III area in the assessment of the air 22 quality relationships. However, a comparison of the median Ecoregion III area S deposition 23 estimates used in the analyses to those used in the aquatic critical load analysis found the 24 difference to typically be less than 7%, with a maximum absolute difference of less than 3 kg/ha-25 yr (as recognized in section 6.3.3 above).

# What do the available information and air quality analyses indicate regarding relationships between air quality metrics based on indicators other than those of the existing standards and S deposition? What are the uncertainties in relationships using such metrics?

Chapter 6 also assessed relationships between co-located measurements and modeled estimates in a subset of Class I areas that are mostly located in the western U.S. The analyses calculated correlations between concentrations of air quality metrics in these locations for indicators other than SO<sub>2</sub> and S deposition in these locations. For example, these results suggest the potential use of IMPROVE PM<sub>2.5</sub>, IMPROVE sulfate, and total sulfur measurements at CASTNET monitoring sites to predict S deposition in those locations. Among those three measurements, concentrations of total S (from SO<sub>2</sub> and SO<sub>4</sub><sup>-2</sup> measurements) at CASTNET sites 1 exhibited the strongest relationship over recent years. These results support the conclusion that S

- 2 deposition in rural areas (such as those in these Class I areas) is mostly comprised of sulfate and
- 3 SO<sub>2</sub>, which is consistent with our understanding of the chemical properties and physical transport
- 4 of these compounds (e.g., that fine particles, such as PM<sub>2.5</sub>, have a much slower dry deposition
- 5 velocity and remain in the atmosphere longer, allowing for transport and deposition in areas
- 6 more distant from sources). Given that this analysis is based on air concentrations and deposition
- 7 estimates at the same locations (distant from sources), use of one of these three combinations of
- 8 S compounds as the indicator of a new standard would entail use of a surveillance network
- 9 designed for this context. Further, a monitoring network for such a standard would also entail
- 10 development of sample collection and analysis FRMs. However, it is unclear whether such an
- 11 approach for a new standard would have advantages over options discussed above.
- 12 7.2.2 N Deposition and Oxides of N and PM

13 To inform conclusions in this review related to the N oxides and PM secondary 14 standards, we consider the information supporting quantitative evaluation of the linkages 15 between N oxides and PM in ambient air with N deposition and associated welfare effects. In 16 considering the questions below, we draw on the available welfare effects evidence described in 17 the current ISA, the 2008 NOx/SOx ISA, the 2009 PM ISA, and past AQCDs, and summarized 18 in chapter 4 above. We do this in combination with the available quantitative information 19 summarized in chapters 5 and 6 above.

### 20 7.2.2.1 Welfare Effects Evidence of Deposition-Related Effects

21 The currently available evidence, including that previously available, documents aquatic 22 and terrestrial effects of N deposition, as summarized in Chapter 4 and described in detail in the 23 ISA. As recognized in section 7.2.1.1 above, N deposition has played a role in acidic deposition 24 in both terrestrial and aquatic ecosystems and associated effects in the U.S. Additionally, the 25 evidence is extensive and longstanding as to the role of N loading of waterbodies and associated 26 eutrophication. Further, the evidence previously available, with noteworthy additions from the 27 more recently available evidence, describes the role of N deposition in terrestrial N enrichment 28 and associated ecosystem effects.

A particular focus of our consideration of the evidence relates to the evidence describing quantitative relationships between deposition and ecosystem effects and the availability of established approaches for estimating risk of such effects from deposition-related N enrichment. The availability of such approaches that can be applied to inform our understanding of spatial extent and magnitude of particular welfare effects associated with different air quality conditions is important to informing decisions on standards that could provide the appropriate control on deposition for the desired level of protection. As recognized in Chapter 5 and section 7.2.1 1 above, the availability of established approaches for quantitatively relating atmospheric

2 deposition to effects on soil and water chemistry and relating those effects to specific welfare

3 effects varies for the different types of ecosystems and categories of effects.

4 We consider here the extent to which such information is available for effects associated 5 with N deposition, and particularly N enrichment-related effects, that might support 6 characterization of the potential for effects, and of the protection that might be afforded for such 7 effects, under different air quality conditions. Our focus with regard to N deposition is on N 8 enrichment-related effects in light of the relatively greater role played by S in acidic deposition, 9 particularly more recently (as described in section 5.2.1.4 above). In so doing, we note the 10 varying directionality of some effects of N enrichment, such that the effects of N enrichment can 11 in particular ecosystems and for particular species seem beneficial (e.g., to growth or survival of 12 those species), although in a multispecies system, effects are more complex with potential for 13 alteration of community composition. Our consideration below of the availability of quantitative 14 information relating atmospheric N deposition to N enrichment-related effects in aquatic and 15 terrestrial ecosystems is in the context of the following question.

# To what extent does the currently available evidence base provide established quantitative approaches for characterizing ecosystem responses to N deposition that can inform judgments on the risk or likelihood of occurrence of ecosystem effects under differing conditions of NO<sub>x</sub> and PM in ambient air?

With regard to acidification-related effects of N deposition, we recognize the approaches and tools referenced in section 7.2.1 above with a focus on S deposition can be utilized for S and N deposition in combination. The approach taken in the analysis of aquatic ecosystem acidification summarized in section 5.2 above was to focus on S deposition, based on analyses indicating the relatively greater role of S deposition under the more recent air quality conditions (as summarized in section 5.2.1.4 above). Discussion of analyses relating acid deposition to terrestrial acidification indicators is also presented in section 5.4 above.

27 With regard to quantitatively analyzing the linkages between N deposition and waterbody 28 eutrophication for the purposes of quantitatively relating N deposition to waterbody responses, 29 we take note of the waterbody-specific nature of such responses and the relative role played by 30 atmospheric deposition. For example, as recognized in the ISA and Chapters 4 and 5 above, the 31 relative contribution to such loading from atmospheric deposition compared to other sources 32 (e.g., agricultural runoff and wastewater discharges), which varies among waterbody types and 33 locations, can be a complicating factor in quantitative analyses. Additionally, characteristics of 34 resident biota populations and other environmental factors are influential in waterbody responses 35 to N loading. Thus, while the evidence is robust as to the ability for N loading to contribute to 36 waterbody eutrophication, which can affect waterbody biota, processes and functions, a variety

of factors complicate our ability to quantitatively relate N deposition rates to eutrophication risks
 in waterbodies ranging from small lakes and streams to large estuaries and coastal waters.

3 With regard to terrestrial ecosystems and effects on trees and other plants, we recognize 4 the complexity, referenced above, that poses challenges to approaches for simulating terrestrial 5 ecosystem responses to N deposition across areas diverse in geography, geology, native 6 vegetation, deposition history and site-specific aspects of other environmental characteristics. 7 Thus, while the evidence is robust as to the ability for N loading from deposition to contribute to 8 changes in plant growth and survival, and associated alterations in terrestrial plant communities, 9 a variety of factors, including the history of deposition and variability of response across the 10 landscape complicate our ability to quantitatively relate specific N deposition rates, associated 11 with various air quality conditions, to N enrichment-related risks of harm to forests and other

12 plant communities in areas across the U.S..

### 13 7.2.2.2 General Approach for Considering Public Welfare Protection

14 As an initial matter, we note that the effects of acidification on plant growth and survival, 15 at the individual level, are generally directionally harmful, including reduced growth and 16 survival. In contrast, the effects of N enrichment can, in particular ecosystems and for particular 17 species, seem beneficial (e.g., to growth or survival of those species), although in a multispecies 18 system, effects are more complex with potential for alteration of community composition. 19 Accordingly, there is added complexity to risk management policy decisions for this category of 20 effects, including the lack of established risk management targets or objectives, particularly in 21 light of historical deposition and its associated effects that have influenced the current status of 22 terrestrial ecosystems, their biota, structure and function.

## What does the currently available quantitative information regarding terrestrial ecosystem responses to N deposition indicate about levels of N deposition that may be associated with increased concern for adverse effects?

26 Focusing first on the evidence for effects of N deposition on trees, we note that the 27 available quantitative information related to effects on plants, including trees, from N deposition 28 summarized in Chapter 5 (and presented in more detail on Appendix 5B) includes soil chemistry 29 modeling analyses for an indicator of soil acidification (as discussed in section 7.2.1 above), as 30 well as studies involving experimental additions of N compounds to defined field plots, and 31 observational studies of potential relationships between tree growth and survival and metrics for 32 N deposition. We consider the latter two types of studies here, as in Chapter 5 above, with regard 33 to what each provides to inform the question posed above. 34 With regard to the information available from experimental addition studies, the ranges

35 of N additions that elicited increased growth overlapped with those that elicited reduced growth

and increased mortality. In considering that these studies were conducted in the context of the natural environment, with a backdrop of the air quality and atmospheric deposition occurring at that time, we note that while some report observations based on additions over just a few years, others extend over a decade or more. In general, they inform our understanding of the effects on tree populations of increased N in forested areas, which can vary, influenced in part by other environmental factors, as well as by species-specific effects on population dynamics. The lowest addition that elicited effects was 15 kg N/ha-yr over a 14-year period occurring from 1988-2002

8 (Appendix 5B, Table 5B-1; McNulty et al., 2005).

9 Among the available observational or gradient studies of N deposition and tree growth 10 and survival (or mortality) are three recently available studies that utilized the USFS/FIA dataset 11 of standardized measurements at sites across the U.S. (Dietze and Moorcroft, 2011; Thomas et 12 al., 2010; Horn et al., 2018). These studies, covering overlapping areas of the U.S. (see Appendix 13 5B, Figure 5B-1), report associations of tree growth and/or survival metrics with various N 14 deposition metrics, providing support to conclusions regarding a role for N deposition in 15 affecting tree health in the U.S., most particularly in regions of the eastern U.S., where 16 confidence in the study associations is greatest (see summaries in section 5.4.2.3 and Appendix 17 5B, section 5B.3.2). The metrics utilized include site-specific estimates of average  $NO_3^{-1}$ 18 deposition and of average total N deposition over three different time periods (Dietze and 19 Moorcroft, 2011; Thomas et al., 2010; Horn et al., 2018). In considering information from these 20 studies below, we note, as recognized in section 5.4.2 and Appendix 5B, the history of N 21 deposition in the eastern U.S. may be an influence in the findings of observational studies, 22 contributing an uncertainty to estimates of a specific magnitude of deposition rate that might be 23 expected to elicit specific tree responses, such as increased or decreased growth or survival.

24 With regard to tree mortality (or survival), the study by Dietze and Moorcroft (2011) 25 reported negative associations of tree mortality with average NO<sub>3</sub><sup>-</sup> deposition (greater survival 26 with greater estimates of  $NO_3^-$  deposition) at sites across the eastern half of the contiguous U.S. 27 The associates were made for plant functional groups comprised of multiple species (Appendix 28 5B, Attachment 1). Site-specific average  $NO_3^-$  deposition in the analysis (1994-2005) ranged 29 from a minimum of 6 kg/ha-yr to a maximum of 16 kg/ha-yr (Dietze and Moorcroft, 2011). At 30 the individual species level, the study by Thomas et al. (2010) reported negative associations of 31 N deposition (mean annual average for 2000-04) with survival (sites of higher deposition had 32 lower survival [higher mortality]) for eight of 23 species in northeastern and north-central U.S 33 and positive associations for three species. Site-specific average N deposition estimates in the 34 analysis (2000-2004) ranged from a minimum of 3 kg/ha-yr to a maximum of 11 kg/ha-yr 35 (Thomas et al., 2010). The other factors analyzed, which included temperature, precipitation and 36 tree size, did not include other pollutants (Thomas et al., 2010). The much larger study by Horn
1 et al. (2018) of 71 species reported associations of tree survival with N deposition that varied

- 2 from positive to negative across the range of deposition at the measurement plots for some
- 3 species, and also varied among species (Appendix 5B, section 5B.3.2.3). Of the six species with
- 4 negative associations of survival with the N deposition metric across the full range of the N
- 5 deposition metric, the median deposition values ranged from 8 to 11 kg N ha ha<sup>-1</sup>yr<sup>-1</sup> (Appendix
- 6 5B, Figure 5B-7). The median deposition values for the 19 other species with hump shape
- 7 functions that were negative at the median deposition value (and for which sample sites were not
- 8 limited to the western U.S.) ranged from 7 to 11 kg N ha<sup>-1</sup>yr<sup>-1</sup>. Values were below 9 kg N/ha-yr

9 for four of the 19 species; these species included at least half of their sample sites in the west or10 in the Northern Forests ecoregion.

11 With regard to growth, the study by Thomas et al. (2010) reported positive associations 12 of N deposition (mean annual average for 2000-04) with tree growth in 11 of 23 species in 13 northeastern and north-central U.S and with negative associations in 3 species. Of the 39 species 14 for which Horn et al (2018) reported significant associations of growth with N deposition, the 15 association was negative across the full deposition range of their sites for two species, pitch pine 16 and bur oak. These species' sites were predominantly in the Atlantic coastal pine barrens and 17 northern plains and forests, respectively. The median deposition across all sites of these species were nine and ten kg N ha<sup>-1</sup>yr<sup>-1</sup> (Appendix 5B, Figure 5B-5). The median deposition values for 18 19 the two other species, with hump shaped functions that were negative at the median, were seven and eight kg N ha<sup>-1</sup>yr<sup>-1</sup>, respectively (Appendix 5B, Figure 5B-5). 20

21 A number of recently available studies have reported on addition experiments involving 22 herb and shrub community response, as summarized in section 5.4.3.1 and Appendix 5B, section 23 5B.3.1. The lowest N additions for which community effects have been reported include 10 kg 24 N/ha-yr. With this addition over a 10-year period, grassland species numbers declined; in a 25 subset of plots for which additions then ceased, relative species umbers increased, converging 26 with controls after 13 years (Appendix 5B, Table 5B-7; Clark and Tilman, 2008). Recent 27 gradient studies of coastal sage scrub in southern California have indicated N deposition above 28 10 or 11 kg/ha-year to be associated with increased risk of conversion to non-native grasslands 29 or reduced species richness (Appendix 5B; section 5B.3.2; Cox et al., 2014; Fenn et al., 2010). A 30 larger observational study considering species richness in open- and closed-canopy communities 31 using database of site assessments conducted over 23-year period and average N deposition 32 estimates for 26-year period reported significant influence of soil pH on the relationship between 33 species richness and N deposition metric. A negative association was observed for low pH 34 forested sites and N deposition above 11.6 kg N/ha-yr (section 5.4.3.1). 35 Observational studies have also analyzed variation in lichen communities in relation to

36 indicators of N deposition as summarized in section 5.4.3.2 and Appendix 5B, section 5B.4.2. In

1 particular, a recent study in the Northwest focused on assessing relationships between metrics for

- 2 community composition and estimated N deposition. In this study the authors identified a
- 3 breakpoint associated with 33-43% fewer oligotrophic species and 3 to 4-fold more eutrophic
- 4 species when total N deposition estimates ranged from 3 to 9 kg N/ha-yr (Geiser et al., 2010).
- 5 Uncertainties associated with these studies include alternate methods for utilizing N deposition
- 6 estimates as well as the potential influence of unaccounted-for environmental factors (e.g.,
- 7 ozone, SO2 and historical air quality and associated deposition), as noted in section 5.4.3.2
- 8 above.

#### 9 7.2.2.3 Relating Deposition-related Effects to Air Quality Metrics

10 As discussed above, in this review, we have explored how well various air quality metrics 11 relate to S and N deposition. The analyses examine, for design value or design value-like 12 metrics, the relationship between measured air quality concentrations and transported S and N 13 deposition. This analysis is particularly relevant given that the current secondary standards are 14 judged using design value metrics based on measurements at the current  $SO_2$ ,  $NO_2$  and  $PM_{2.5}$ 15 FRM/FEM monitors. This information can help inform how changes in NO<sub>2</sub> emissions relate to 16 changes in deposition and how best to regulate measured air quality concentrations through the 17 NAAQS to maintain N deposition at or below certain levels. The details of these analyses are 18 described in Chapter 6 and Appendices 2A and 6A. In addressing the questions below, we 19 consider the findings of those analyses specific to N deposition associated with N oxides and 20 PM.

#### 21 22 23

### • What does the available information and air quality analyses indicate regarding relationships between air quality metrics related to the existing standards and N deposition? What are the uncertainties in relationships using such metrics?

For N, the results in Chapter 6 suggest that oxidized N deposition in rural areas is mostly from deposition of nitric acid and particulate nitrate, rather than NO<sub>2</sub>. Additionally, the results suggest that in some areas inorganic nitrogen (e.g., NH<sub>4</sub>) contributes to the N deposition, with higher contributions in areas near emission sources of NH<sub>3</sub>.

28 In considering policy options that might be expected to provide the desired protection of 29 the public welfare from N deposition related effects, we consider the current form and averaging 30 time of the secondary NO<sub>2</sub> NAAQS which is the annual average of NO<sub>2</sub>. As in the assessments 31 of the other pollutants and air quality metrics, the analyses focus on a 3-year average for  $NO_2$ 32 and N deposition and include multiple years of data to better assess more typical relationships. 33 For NO<sub>2</sub>, the correlations between annual average  $NO_2$  and N deposition in the analyses that 34 considered transport were somewhat low (as described in section 6.2.2 above), indicating some 35 uncertainty in the extent to which a standard set as this air quality metric might control N

1 deposition by itself. However, the analyses also found that the correlation between annual 2 average  $PM_{2.5}$  and N deposition was much stronger, likely due to  $HNO_3$ ,  $NO_3$  and  $NH_4$  being the 3 largest contributors to N deposition and being most closely related to concentrations of PM<sub>2.5</sub>. 4 Given this information and these relationships, the results suggest the potential for a standard set 5 as the PM<sub>2.5</sub> annual average, averaged over three years, to better control N deposition. Such a 6 metric would also provide some control over S deposition, as discussed above. Of course, it is 7 important to also keep in mind that PM2.5 monitors that contribute to the S and N deposition 8 across the U.S. also measure other non-S and N related pollutants as part of the  $PM_{2.5}$  total mass. 9 This and other uncertainties in the analyses are noted in Chapter 6. However, it is unclear how

10 much and in what way each of these uncertainties might impact the results.

# What does the available information and air quality analyses indicate regarding relationships between air quality metrics based on indicators other than those of the existing standards and N deposition? What are the uncertainties in relationships using such metrics?

15 As discussed above, Chapter 6 also assessed relationships between co-located 16 measurements and modeled estimates in a subset of Class I areas that are mostly located in the 17 western U.S. The analyses indicated correlations between concentrations of other air quality 18 metrics and N deposition levels in these locations. For example, these results suggest that N 19 deposition in these rural areas is fairly well correlated with air concentrations of nitric acid and 20 particulate nitrate. Additionally, the results suggest that IMPROVE PM2.5, IMPROVE approximated inorganic N PM<sub>2.5</sub> (NO<sub>3</sub><sup>-</sup> + NH<sub>4</sub><sup>+</sup>,  $\mu$ g/m<sup>-3</sup>), and inorganic nitrogen measured at 21 CASTNET monitoring sites (HNO<sub>3</sub> + NO<sub>3</sub><sup>-</sup> + NH<sub>4</sub><sup>+</sup>,  $\mu$ g/m<sup>-3</sup>) can be used to predict N deposition 22 23 in these locations, with CASTNET N showing the most consistent relationship over recent years 24 (section 6.2.1.4).

25 As similarly discussed above for S, given that this analysis is based on air concentrations 26 and deposition estimates at the same locations (distant from sources), use of one or more of these 27 air quality metrics as the indicator of a new standard would entail use of a surveillance network; 28 attainment of such a standard would then be judged based on these monitor measurements in 29 these Class I or other similar locations using monitoring networks like CASTNET and/or 30 IMPROVE (e.g., with locations generally in rural areas, away from sources). A corresponding 31 FRM/FEM would need to be developed for these monitors and measurements and adequacy of 32 network coverage would need to be assessed. However, it is unclear whether there are 33 advantages to such a choice for a new or revised standard versus those options discussed above.

#### 1 7.3 PRELIMINARY CONCLUSIONS

This section describes preliminary conclusions for the Administrator's consideration in this review of the secondary NAAQS for oxides of N and S and for PM standards. These conclusions are based on consideration of the assessment and integrative synthesis of the evidence (as summarized in the ISA, and the 2008 ISA and AQCDs from prior reviews), and the quantitative information on exposure and air quality summarized above. Taking into consideration the discussions above in this chapter, this section addresses the following overarching policy question.

## Do the current evidence and quantitative analyses call into question the adequacy of protection from ecological effects afforded by the SO<sub>2</sub>, NO<sub>2</sub> and PM secondary standards? What alternate standards may be appropriate to consider with regard to protection from ecological effects of SOx, NOx and PM?

13 In considering this question, we first recognize what the CAA specifies with regard to 14 protection to be provided by the secondary standards. Under section 109(b)(2) of the CAA, the 15 secondary standard is to "specify a level of air quality the attainment and maintenance of which 16 in the judgment of the Administrator ... is requisite to protect the public welfare from any known 17 or anticipated adverse effects associated with the presence of such air pollutant in the ambient 18 air." The secondary standard is not meant to protect against all known or anticipated SO<sub>2</sub> related 19 welfare effects, but rather those that are judged to be adverse to the public welfare, and a bright-20 line determination of adversity is not required in judging what is requisite (78 FR 3212, January 21 15, 2013; 80 FR 65376, October 26, 2015; see also 73 FR 16496, March 27, 2008). Thus, our 22 consideration of the currently available information regarding welfare effects of the oxides of 23 sulfur and nitrogen and of PM is in this context, while recognizing that the level of protection 24 from known or anticipated adverse effects to public welfare that is requisite for the secondary 25 standard is a public welfare policy judgment to be made by the Administrator. 26 The general approach in a review of a secondary NAAQS, and accordingly in PAs, 27 involves, first, evaluation of the currently available information with regard to key considerations

for assessing risk of or protection against the effects of the criteria pollutant of focus, such as discussed in section 3.4 above. In this evaluation, the PA considers the welfare effects of the

- discussed in section 3.4 above. In this evaluation, the PA considers the welfare effects of the pollutant, associated public welfare implications, and also the quantitative information, such as
- 31 regarding exposure-response relationships, and associated tools or metrics, as well as associated
- 32 limitations and uncertainties. The quantitative tools (e.g., metrics for effects and metrics for
- 33 summarizing exposures) allow for identification and assessment of exposures of concern and,
- 34 correspondingly, of exposures appropriate for focus in assessing protection afforded by the
- 35 existing standard, and as appropriate, in assessing potential alternatives. The latter part of the
- 36 general approach in a review and a PA is then consideration of the extent to which the existing

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1 standard provides air quality that would be expected to achieve such protection and, as

- 2 appropriate, potential alternative options (standard or standards) that could be expected to
- 3 achieve this desired air quality. This consideration goes beyond a focus on the key exposure
- 4 metrics and concentrations of potential concern to whether the form, averaging time and level of
- 5 the standard (or suite of standards), together, provide the requisite protection.
- 6 In NAAQS reviews in general, the extent to which the protection provided by the current 7 secondary standards for oxides of S and N and for PM are judged to be adequate depends on a
- 8 variety of factors, including science policy judgments and public welfare policy judgments.
- 9 These factors include public welfare policy judgments concerning the appropriate benchmarks
- 10 on which to place weight, as well as judgments on the public welfare significance of the effects
- 11 that have been observed at the exposures evaluated in the welfare effects evidence. The factors
- 12 relevant to judging the adequacy of the standard also include the interpretation of, and decisions
- 13 as to the weight to place on, different aspects of the quantitative analyses of air quality and
- 14 exposure and any associated uncertainties. Additionally, to the extent multiple policy options are
- 15 identified that would be expected to achieve a desired level of protection, decisions on the
- 16 approach to adopt falls within the scope of the Administrator's judgment. Thus, we recognize
- 17 that the Administrator's conclusions regarding the adequacy of the current standard will depend
- 18 in part on public welfare policy judgments, on science policy judgments regarding aspects of the
- 19 evidence and exposure/risk estimates, and on judgments about the level of public welfare
- 20 protection that is requisite under the Clean Air Act.
- 21 In the discussion below we address first the SO<sub>2</sub> standard, and its adequacy with regard to 22 protection of the public welfare from the direct effects of SOx in ambient air. Next, we address 23 the extent of protection provided by the SO<sub>2</sub> standard from deposition related effects of SOx in 24 ambient air, and consideration of alternate standards for this purpose. In so doing, we focus 25 primarily on the contribution of SOx in ambient air to ecosystem acidification and particularly 26 aquatic acidification. After addressing SOx in this way, we next consider the NO2 standard and 27 its adequacy with regard to protection of the public welfare from direct effects of N oxides in 28 ambient air, as well as the extent of protection provided by the NO<sub>2</sub> standard from deposition 29 related effects of N oxides in ambient air and consideration of alternate standards for this 30 purpose. Lastly, we address the PM standards and the extent of their protection of the public 31 welfare from ecological effects. In each of these discussions, we recognize limitations in the 32 available information and tools and associated uncertainties, which we recognize to vary in 33 specificity and significance.
- As noted earlier in this draft PA, the SO<sub>2</sub> secondary standard is 0.5 ppm, as a 3-hour average concentration not to be exceeded more than once per year. The evidence of welfare effects at the time this standard was established in 1971 indicated the effects of SOx on

1 vegetation, most particularly effects on foliar surfaces. The currently available information

- 2 continues to document the occurrence of visible foliar injury as a result of acute/short (a few
- 3 hours) exposure, with greater exposures (repeated and/or of longer duration) affecting plant
- 4 growth and yield. As summarized in the ISA, there is "no clear evidence of acute foliar injury
- 5 below the level of the current standard" (ISA, section IS.4.1, P. IS-37). We additionally note that
- 6 across all sites meeting the existing standard (outside Hawaii, where air quality can be influenced
- 7 by volcanic emissions) during all years since 2000, 95% of the maximum annual 3-hour average
- 8 concentrations are below 0.2 ppm and 99% are below 0.3 ppm. Thus, we find that the currently
- 9 available information, including that newly available in this review, does not call into question
- 10 the adequacy of protection provided by the existing  $SO_2$  standard from the direct effects of SOx
- 11 in ambient. Accordingly, we conclude that it is appropriate to consider retaining the existing
- 12 standard for that purpose. In so doing, however we recognize the extensive evidence of the
- 13 contribution of SOx in ambient air to acidic deposition, particularly in aquatic ecosystems and
- we next consider the adequacy of protection afforded by the existing SO<sub>2</sub> standard from sucheffects.
- 16 With regard to deposition-related effects, we note the range of median deposition values 17 estimated across U.S. ecoregions (in terms of level 3 specification) for the 20-year period 18 analyzed (2001-2020) extended up through 10 kg S/ha-yr to as high as 20 kg S/ha-yr during 19 years when the existing SO<sub>2</sub> standard was met, and when design values for the standard (second 20 highest 3-hour average in a year) ranged below 500 ppb (as discussed in section 6.2.2.2 above). 21 For example, in the earliest 3-yr period (2001-03), when some ecoregion max DV values ranged below 400 ppb median S deposition in 4 ecoregions exceeded 15 kg/ha<sup>-1</sup>vr<sup>-1</sup> and median S 22 23 deposition in more than 10 ecoregions exceeded 10-12 kg/ha<sup>-1</sup>yr<sup>-1</sup>.
- Considering the aquatic acidification estimates of S deposition on the order of 12-15 kg S/ha-yr levels occurring during periods when the existing standard has been met, and the aquatic acidification analysis results for that level of S deposition, it is reasonably concluded that the current evidence and quantitative analyses call into question the adequacy of the existing standard. Thus, we have evaluated options for potential alternative standards that may be indicated to provide appropriate control of S deposition and associated welfare effects.
- For the purposes of evaluating options for potential alternative standards that may be considered to provide an appropriate level of protection from deposition-related effects of S oxides, we have drawn on the quantitative analyses and information described in Chapter 5 and summarized in section 7.2.1.2 above. In this context and for our purposes within this PA, we have focused on a range of S deposition levels below 12 kg S/ha-yr, extending down as low as 4 or 5 kg S/ha-yr. In focusing on this range, we draw primarily from the aquatic acidification analyses. In so doing, however, we also note the linkages between watershed soils and

1 waterbody acidification, as well as terrestrial effects. Such linkages indicate that a focus on 2 protecting waterbodies from reduced ANC will also, necessarily, provide protection for 3 watershed soils, and may reasonably be expected to also contribute protection for terrestrial 4 effects. That notwithstanding, we recognize there to be limitations of the quantitative analyses 5 and associated uncertainties in their interpretation, as referenced in Chapter 5. Accordingly, in 6 focusing on this range for our purposes here, we note there to be relatively greater uncertainty 7 associated with the lower levels. Moreover, we recognize that, in the end, judgments inherent in 8 identification of such a range, include judgements related to the weighing of uncertainties, as 9 well as the consideration of the appropriate targets for public welfare protection, and fall within 10 the purview of the Administrator.

11 In considering options for a secondary standard that might be concluded to provide the 12 desired control of S deposition, we first note the complexity of identifying a national air quality 13 standard focused on protection from national deposition patterns (rather than air concentrations) 14 of concern to the public welfare. For example, atmospheric deposition (ecosystem loading) of S, 15 is, in a simple sense, the product of atmospheric concentrations of S compounds, factors 16 affecting S transfer from air to surfaces, and time. Further, atmospheric concentrations in an 17 ecosystem are, themselves, the result of emissions from multiple, distributed sources, 18 atmospheric chemistry and transport. Accordingly, consideration of the location of source 19 emissions and expected pollutant transport (in addition to the influence of physical and chemical 20 processes) is essential to identification of options expected to provide a particular level of 21 deposition control in sensitive ecosystems. Further, we recognize that to achieve the desired level 22 of S deposition control in sensitive ecosystems, SO<sub>2</sub> emissions must be controlled at their 23 sources and that such control can be provided by the appropriate secondary standard measured at 24 regulatory SO<sub>2</sub> monitors given that these monitors are generally sited near large SO<sub>2</sub> sources to 25 provide control of these large sources of SO<sub>2</sub>. While recognizing the variation across the U.S. in 26 the processes that govern that transformation of source emissions to eventual deposition of S 27 compounds, we utilized a trajectory-based approach to account for the relationship between 28 upwind concentrations near sources and deposition in areas more distant, as described in section 29 6.2.2 above. Based on application of this approach, we observed that while there is some 30 variation (especially at lower deposition levels), there is generally a strong positive linear correlation between deposition estimates and the trajectory-based concentration metrics. While 31 32 there is residual uncertainty in the relationship, its use facilitates the linking of pollutant 33 concentrations and the resultant N or S deposition, with the deposition-related welfare effects 34 associated with various deposition levels. With this linkage, the protectiveness of existing 35 standards can then be considered in terms of pollutant concentrations.

1 Based on the analyses described in the preceding chapters and all of the above 2 considerations, we have identified options for potential alternative SO<sub>2</sub> standards appropriate to 3 consider for providing control of S deposition associated with SOx in ambient air. The potential 4 alternatives include a SO<sub>2</sub> standard of the same averaging time and form as the existing 5 secondary standard, with a level revised to within a range of 200 to 400 ppb. Additionally, 6 however, we recognize, in light of the 'not to be exceeded more than once per year' form of the 7 existing standard, which allows an average concentration above the standard level for which 8 there is no limit, and the relatively short averaging time, that this option might reasonably be 9 considered a relatively imprecise approach for controlling S deposition. Accordingly, we 10 conclude it may be more appropriate to consider adoption of a new SO<sub>2</sub> standard with a different 11 averaging time and form, as well as level, such as a standard with an averaging time of one year, 12 and a form of the average of annual averages across three consecutive years. For such a standard, 13 based on the air quality analyses and recognizing the various limitations and associated 14 uncertainties, a level on the order of 22 ppb to 10 ppb is identified. We additionally note that 15 whether such a range is concluded to be appropriate and/or what value within this range of levels 16 might be appropriate, are in the end decisions made by the Administrator, in light of judgments 17 associated with weighing of the differing aspects of the evidence and air quality information and 18 how to consider their associated uncertainties and limitations.

19 Turning to consideration of the secondary standard for oxides of N, we note that the 20 existing secondary standard for oxides of N is 53 ppb, as an annual mean. The evidence of 21 welfare effects at the time this standard was established in 1971 indicated the direct effects of N 22 oxides on vegetation, most particularly effects on foliar surfaces. The currently available 23 information continues to document such effects, as summarized in sections 4.1 and 5.1.2 above. 24 We also recognize the evidence of  $NO_2$  concentrations well in excess of the standard that occurred for more than a decade in areas of California where damage suspected to relate to N 25 26 oxides in air is well documented (as summarized in sections 5.1.2, 5.4.3 and Appendix 5B, 27 sections 5B.4). Given the extensive period of elevated concentrations above the standard, the 28 evidence is not clear as to the potential for such effects to have been elicited by air quality that 29 met the standard. Thus, the available information while clearly documenting the potential for N 30 oxides in ambient air to cause harm is not clear as to the extent to which it may call into question 31 or support the adequacy of protection provided by the current NO<sub>2</sub> standard. The experimental 32 evidence also does not provide clear indication of welfare effects associated with exposure 33 concentrations that might be allowed by the current standard. We note, however, that depending 34 on judgments as to the weight to place on specific aspect aspects of the evidence and air quality 35 analyses, and associated uncertainties, it may be judged appropriate to consider a more restrictive 36 NO<sub>2</sub> standard that might be considered to offer the potential for some desired additional

1 protection from deposition-related ecosystem effects, and also the potential for increased 2 protection from effects related to airborne nitric acid effects on biota surfaces. Accordingly, in 3 addition to preliminarily concluding it is appropriate to consider retaining the existing NO<sub>2</sub> 4 standard, we additionally identify a revision option for the secondary standard for N oxides. 5 In considering options for revision of the secondary standard for N oxides, we have 6 further evaluated the information related to deposition-related effects on ecosystems. With regard 7 to the currently available information related to deposition -related effects of N oxides on 8 ecosystems, we recognize, as discussed in section 7.2.2 above, the complexities and challenges 9 associated with quantitative characterization of N enrichment-related effects in terrestrial or 10 aquatic ecosystems across the U.S. that might be expected to occur due to specific rates of 11 atmospheric deposition of N over prolonged periods, and of the associated uncertainty. Some 12 complications with regard to terrestrial deposition are similar to those for aquatic deposition, 13 such as untangling the impacts of historic deposition from what might be expected from specific 14 annual deposition rates absent that history, while others, related to available quantitative 15 information and analyses, differ. Further, with regard to many aquatic systems, for which there 16 are non-air contributing sources, we recognize the complexity of estimating the portion of N 17 inputs, and associated contribution to effects, derived from atmospheric sources. Lastly, as noted 18 above, there is additional complexity in risk management policy decisions for this category of 19 effects, including with regard to risk management targets or objectives for an ecosystem stressor 20 like N enrichment, particularly in light of historical deposition and its associated effects that have 21 influenced the current status of terrestrial ecosystems, their biota, structure and function.

22 Additionally, as discussed in section 7.2.2.3 above, and in more detail in Chapter 6, 23 several observations are made based on the air quality analyses of relationships between N 24 deposition and NO<sub>2</sub> and PM<sub>2.5</sub> air quality metrics. For NO<sub>2</sub>, the correlations between annual 25 average NO2 and N deposition in the analyses that considered transport were somewhat low (as 26 described in section 6.2.2 above), indicating some uncertainty in the extent to which a standard 27 set as this air quality metric might control N deposition by itself. These analyses found a much 28 stronger correlation between annual average PM<sub>2.5</sub> and N deposition. This finding is likely due to 29 HNO<sub>3</sub>, NO<sub>3</sub> and NH<sub>4</sub> being the largest contributors to N deposition and also being most closely 30 related to concentrations of PM<sub>2.5</sub>. Given this information and these relationships, the results 31 suggest the potential for a standard set as the PM<sub>2.5</sub> annual average, averaged over three years, to 32 be a better air quality metric for control N deposition than the NO<sub>2</sub> metric assessed. Such a 33 metric would also be expected to provide some control of S deposition, as discussed above. In 34 recognizing this finding, however, we also note that PM<sub>2.5</sub> across the U.S. varies with regard to 35 composition, including the contribution from other pollutants that are not S or N containing. The potential influence of this and other uncertainties in the analyses (noted in Chapter 6) is unclear. 36

1 Thus, based on the current evidence and quantitative air quality, exposure and risk 2 information, with associated limitations and uncertainties, in light of all of the considerations 3 above, we preliminarily conclude it is appropriate for the Administrator to consider an array of 4 policy options supported by the current scientific information and quantitative air quality, 5 exposure and risk analyses. The potential policy options that could inform the Administrator's 6 decisions on the NAAQS providing the "requisite" public welfare protection and that are 7 supported by the science include both options to address protection of direct effects of the 8 pollutants in ambient air and options to address protection of effects related to S deposition and 9 related to N deposition. A summary of these options is shown in Table 7-1 and described below.

10 To address protection of the public welfare from effects of SOx in ambient air, we 11 recognize options appropriate to consider for protection from both direct and deposition-related 12 effects. With regard to protection against the direct effects of SOx in ambient air, we conclude it 13 is appropriate to consider retaining the current secondary standard. To address protection of the 14 public welfare from effects related to S deposition, we have identified two options, one involving 15 revisions to the existing SO<sub>2</sub> standard to additionally afford protection for S deposition-related 16 welfare effects, and one involving adoption of a new SO<sub>2</sub> standard. The option involving revision 17 of the existing standard is for a standard with a level revised to 200-400 ppb, as the 2nd highest 18 daily 3-hour maximum, and a form revised to be the average over three consecutive years. An 19 alternate option is to establish an additional SO<sub>2</sub> annual mean standard, averaged across three 20 years, with a level within the range from 22 to 10 ppb, with greater uncertainty for lower levels. 21 With this option, it may be appropriate to consider revoking the current 3-hour standard.

With regard to protection from effects of N oxides and N deposition, three options are identified in consideration of: limitations in the available evidence, and associated uncertainties related to interpretation of the evidence and air quality information; relationships between the two pollutants and associated effects; and connections of effects elicited by N oxides in ambient air and deposited onto biota surfaces.

27 One option is based on judgments that the evidence for direct effects of N oxides and PM 28 does not call into question the adequacy of protection provided by these standards and also 29 judgments that weigh heavily the limitations, and associated uncertainties associated with the 30 evidence base for ecosystem effects related to N deposition, such as N enrichment, as discussed 31 above, and with the air quality information related to the potential for control of N deposition in 32 areas across the U.S., in light of variation in the composition of both oxides of N and of PM. The 33 first set of limitations and uncertainties relates to quantitative relationships between N deposition 34 and ecosystem effects, based on which differing judgments may be made in decisions regarding 35 protection of the public welfare. In the case of protection of the public welfare from adverse 36 effects associated with nutrient enrichment, we additionally recognize the complexity associated

7-29

1 with identification of appropriate protection objectives in the context of changing conditions in

2 aquatic and terrestrial systems as recent deposition has declined from the historical rates of

- 3 loading. The second set of limitations and uncertainties relates to relatively lower correlations of
- 4 air quality metrics for N oxides with N deposition in ecosystems and the variation in PM
- 5 composition across the U.S., particularly that between the eastern and western U.S. The option
- 6 based on all of these considerations is to retain the existing NO<sub>2</sub> and PM standards based on the
- 7 judgment that the current evidence does not call into question the adequacy of protection of the
- 8 public welfare from both direct effects of N oxides and PM in ambient air and effects related to
- 9 N deposition.

10 To the extent different judgments are made, two options for revision are identified that 11 might be expected to provide protection from both direct effects of N oxides in ambient air and 12 from N deposition of potential concern. Based on the air quality information that suggests better 13 control of N-deposition with the annual PM2.5 versus NO2 standard, this option involves revision 14 to the level of the PM<sub>2.5</sub> annual secondary standard. For this option, it may be appropriate to 15 consider revisions to the level of the current PM<sub>2.5</sub> (annual) standard of 15  $\mu$ g/m<sup>3</sup> down to a level 16 as low as  $12 \,\mu \text{g/m}^3$ , recognizing increased uncertainty associated with lower levels. Such a 17 standard would potentially provide additional protection against S deposition. A second option 18 for revision is recognized, taking into account limitations in the available evidence, and 19 associated uncertainties related to interpretation of the evidence of terrestrial biota effects of 20 nitric acid, which may be the direct effects most sensitive to oxides of N in ambient air. We note 21 that such effects may be considered to be both direct effects and also deposition-related effects as 22 they relate to direct contact with biota surfaces by dry deposition (e.g., ISA, Appendix 3, section 23 3.4, Appendix 5, section 5.2.3 and Appendix 6, section 6.3.7). The options include retaining or 24 revising the current secondary  $NO_2$  standard. For the revision option, it may be appropriate to 25 consider levels below 53 ppb. In considering such lower levels, potentially extending down to 26 perhaps, as low as about 40 ppb, however, we recognize appreciably greater uncertainty with 27 decreasing levels below 53 ppb.

28 In addition to the options identified above, we additionally recognize the potential value 29 in establishment of a revised standard or suite of standards with alternate indicator(s) that may 30 target specific chemicals that deposit N and S (e.g., NO<sub>3</sub>, SO<sub>4</sub>, NH<sub>4</sub>). In so doing, we note a 31 number of information gaps that would need to be filled to inform identification of specific 32 options of this type. One example relates to the depth of our understanding of the distribution of 33 these chemicals in ambient air, including relationships between concentrations near sources and 34 in areas of deposition, such as protected areas. In this context we recognize that depending on the 35 indicator selected the relationship exhibited between concentrations of the indicator and N or S deposition at the same location may not be expected to hold for concentrations of the indicator in 36

1 more distant locations, including locations near emissions sources. Additionally, we recognize

- 2 the practical considerations associated with establishing new standards with new indicators
- 3 related to establishment of regulatory measurement methods and surveillance networks, that
- 4 would yield effective implementation of the standards. Thus, while we note the potential value in
- 5 such approaches, we also recognize the additional data collection and analysis needed to support
- 6 their adoption.

7 We additionally note that the Administrator's decisions regarding secondary standards, in 8 general, are largely public welfare judgments, as described above. We note that different public 9 welfare policy judgments could lead to different conclusions regarding the extent to which the 10 current and various alternative standards might be expected to provide the requisite protection of 11 the public welfare. Such public welfare judgments include those related to identification of 12 effects of public welfare significance, as well as with regard to the appropriate weight to be 13 given to differing aspects of the evidence and air quality information, and how to consider their 14 associated uncertainties and limitations. For example, different judgments might give greater 15 weight to more uncertain aspects of the evidence. There are additionally, judgments with regard 16 to the appropriate objectives for the requisite protection of the public welfare. Such judgments 17 are left to the discretion of the Administrator. Thus, in identifying a broad array of options for 18 consideration, we note that decisions on the approach to take in achieving the desired air quality 19 and public welfare protection fall within the scope of the Administrator's judgment. 20

Current Standards Protect against Direct Effects of Pollutants in Ambient Air					
Indicator		Level	Form	Avg Time	
SO <sub>2</sub>		0.5 ppm	Not to be exceeded more than once per year	3 hours	
NO <sub>2</sub>		53 ppb	Annual	1 year	
DM.		15 µg/m³	Annual, averaged over 3 years	1 year	
PM2.5		35 µg/m³	98 <sup>th</sup> percentile, averaged over 3 years	24 hours	
PM <sub>10</sub>		150 µg/m³	Not to be exceeded more than once per year on average over 3 years	24 hours	
	Draft Policy Ass	sessment Range	of Options for Consideration		
Retain/Rev	vise	Revise to Addre	ess Deposition-related Effects		
to Address	Direct Air-related Effects	Level	Form	Avg Time	
	Retain	200-400 ppb	Not to be exceeded more than once per year, averaged over 3-years	3 hours	
SO <sub>2</sub>		OR:			
		10-22 ppb	Annual, averaged over 3 years	1 year	
	Retain <b>OR</b>				
NO <sub>2</sub>	Revise level of annual NO <sub>2</sub> s	tandard to <53 ppl	to <53 ppb to as low as 40 ppb		
		Provide increased protection using a revised PM <sub>2.5</sub> standard			
PM <sub>2.5</sub> annual	Retain	<15 µg/m³ to as low as 12 µg/m³	Annual, averaged over 3 years	1 year	
standard		OR Retain			
PM <sub>2.5</sub> 24-hour standard PM <sub>10</sub> standards	Retain	Not assessed as	most relevant metrics for N and S deposi	tion	
We additionally recognize the potential for establishment of a revised standard or suite of standards with alternate indicator(s) that may target specific chemicals that deposit N and S (e.g., NO <sub>3</sub> , SO <sub>4</sub> , NH <sub>4</sub> ). A number of uncertainties and complications are recognized with this option that include uncertainties in relationships between concentrations near sources and in areas of deposition, as well as additional time and resources related to establishment of regulatory monitoring networks and measurement methods.					

### Table 7-1. Summary of current standards and draft range of potential policy options for consideration.

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#### 7.4 AREAS FOR FUTURE RESEARCH RELATED TO KEY **UNCERTAINTIES AND**

3 In this section, we highlight key uncertainties associated with reviewing and establishing 4 the secondary standards for oxides of S, oxides of N and PM, and additionally recognize that 5 research in these areas may additionally be informative to the development of more efficient and 6 effective control strategies. Accordingly, areas highlighted for future welfare effects and 7 atmospheric chemistry research include model development, and data collection activities to 8 address key uncertainties and limitations in the current scientific evidence. These areas are 9 similar to those highlighted in past reviews, such as those that follow: 10 • Data and tools to relate concentrations of specific pollutants in ambient air with

- 11 deposition. This could include expansion of existing monitoring networks (either in 12 number or in the number of pollutants measured) to enable more geographically 13 representative comparisons of local deposition and local air quality concentrations.
- 14 • Research to further develop and improve modeling tools that relate atmospheric 15 deposition of specific compounds to changes in soil conditions, which influence watershed aquatic impacts as well as effects on resident vegetation, in areas characterized 16 by different soil types and geology. 17
- 18 • Continued refinement of the TDEP methodology to estimate national total deposition. 19 This could include efforts to continually evaluate and improve the air quality model 20 simulation inputs to TDEP.
- 21 • Additional work to improve accuracy of estimates of BCw, a critical parameter in modeling to characterize risks associated with aquatic and terrestrial acidification. 22
- 23 • To address uncertainty associated with characterizing risks associated with terrestrial 24 acidification, additional research might contributed to an improved understanding of 25 effects on sensitive vegetation of various levels of BC:Al in different soil types.
- 26 • Improved understanding or relationship between soil N and C:N metrics and effects on 27 key ecological receptors.
- 28 • Although addition or exposure studies are somewhat limited, studies assessing important 29 tree species included in Horn et al 2018 would help improve confidence.
- 30 • Research to improve understanding of the linkages between deposition, geochemical 31 metrics and ecological effects of freshwater ecosystem eutrophication. Currently 32 available studies of waterbodies in the western U.S. have included investigations of 33 nutrient limitation and diatom assemblages. Studies in eastern lakes and streams have 34 primarily focused on NO3 leaching. Information is limited for relationships between 35 additional ecological endpoints (e.g., effects on fish and invertebrate communities) and NO3 concentrations (or other chemical indicators). 36
- 37 • Research relating specific indicators of acidification or nutrient enrichment to ecological 38 effects and to ecosystem services (e.g., fish harvest, recreation, etc).

- Research to address key limitations and uncertainties in modeling watershed N loading,
   including atmospheric deposition to indicators of eutrophication (e.g., disolved oxygen
   and chlorophyll A). For example, data to better estimate estuary-specific parameters (e.g.,
   as used in Evans and Scavia Model); improved modeling tools that combine watershed
   loading and influence on estuarine indicators.
- Information is limited relating N deposition to specific endpoints in wetlands. Additional research would contribute to an improved understanding of relationships between N deposition and chemical and ecological responses across a range of wetland types and across geographic regions.
- Regarding aquatic eutrophication, research in several areas would advance assessment approaches. These include research on appropriate endpoints or indicators; important mediating factors (e.g., drought, temperatures, seasonality, DOC, recovery from acidification) and characterization of their role in key processes, as well as on the extent of differences among N compounds with regard to their role in key processes.

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32		

#### **5A.1 AQUATIC ACIDIFICATION AND OVERVIEW OF ANALYSES**

2 Air emissions of sulfur oxides  $(SO_x)$ , nitrogen oxides  $(NO_x)$ , and reduced forms of 3 nitrogen (NH<sub>x</sub>) react in the atmosphere through a complex mix of reactions and thermodynamic 4 processes in gaseous, liquid, and solid phases to form various acidifying compounds. These 5 compounds are removed from the atmosphere through wet (e.g., rain, snow), cloud and fog, or dry (e.g., gases, particles) deposition. Deposition of SO<sub>x</sub>, NO<sub>x</sub>, and NH<sub>x</sub> leads to ecosystem 6 7 exposure to acidification. The 2020 ISA concludes that the body of evidence is sufficient to infer 8 a causal relationship between acidifying deposition and adverse changes in freshwater biota (see 9 ISA, Appendix 8). Freshwater systems of the U.S. include lakes, rivers, streams, and wetlands. 10 Changes in biogeochemical processes and water chemistry caused by deposition of N and S to 11 surface waters and their watersheds have been well characterized for decades and have 12 ramifications for biological functioning of freshwater ecosystems. 13 When S or N deposition leaches from soils to surface waters in the form of sulfate  $(SO_4^{2^-})$ 14 or nitrate ( $NO_3^{-}$ ), an equivalent number of positive cations, or countercharge, is also transported. 15 This maintains electroneutrality. If the countercharge is provided by base cations such as calcium

- 16 (Ca<sup>2+</sup>), magnesium (Mg<sup>2+</sup>), sodium (Na<sup>+</sup>), or potassium (K<sup>+</sup>), rather than hydrogen (H<sup>+</sup>) and
- 17 aluminum (Al<sub>3</sub><sup>+</sup>), the acidity of the soil water is neutralized, but the base saturation of the soil is
- 18 reduced. Continued  $SO_4^{2-}$  and/or  $NO_3^{-}$  leaching can deplete available base cation pools in the
- 19 soil. As the base cations are removed, continued deposition and leaching of  $SO_4^{2-}$  and/or  $NO_3^{-}$  -
- 20 (with  $H^+$  and  $Al_3^+$ ) leads to acidification of soil water, and by connection, surface water. Loss of
- soil base saturation is a cumulative effect that increases the sensitivity of the watershed to furtheracidifying deposition.
- 23 These chemical changes in water quality can occur over both long- and short-term 24 timescales. Short-term (i.e., hours or days), often termed episodic, periods of increased acidity 25 can also have significant biological effects. Episodic chemistry refers to conditions during 26 precipitation or snowmelt events when proportionately more drainage water is routed through 27 upper soil horizons that tends to provide less acid neutralizing than deeper soil horizons. Surface 28 water chemistry has lower pH and acid neutralizing capacity (ANC) during these events than 29 during baseflow conditions. Acid neutralizing capacity is defined as the total amount of strong 30 base ions minus the total amount of strong acid anions as the differences between the equivalent sum of base cations (SBC) plus ammonium ( $Ca^{2+} + Mg^{2+} + K^+ + Na^+ + NH_4^+$ ) and the equivalent 31 sum of acid anions (SAA)  $(SO_4^{2-} + NO_3^{-} + Cl^{-})$  (eqn. 5A-1): 32

33 
$$ANC = SBC - SAA = (Ca^{2+} + Mg^{2+} + K^{+} + Na^{+} + NH_{4}^{+}) - (SO_{4}^{2-} + NO_{3}^{-} + Cl^{-})$$
 (5A-1)

5A-1

1 Acid neutralizing capacity and pH are related to one another as they both are measures of 2 acidity in surface waters and low pH values correspond to low ANC values. However, pH in 3 natural waters is dependent on the amount of carbon dioxide, organic acids, and aluminum 4 solubility, which impacts the relationships between the two parameters. The amount of carbon 5 dioxide (CO<sub>2</sub>) dissolved in surface waters is affected by biological activity and temperature, 6 which decreases pH but does not impact ANC. Dissolved organic carbon (DOC), which includes 7 organic acids (e.g., fulvic and humic acids, carboxylic acids, and amino acids), also lowers pH 8 values in surface waters and changes the relationship between pH and ANC (ISA, Appendix 4, 9 section 4.3.9).

10 The principal factor governing the sensitivity of aquatic ecosystems to acidification from 11 acidifying deposition is geology [particularly surficial geology; (Greaver et al., 2012)]. Levels of 12 acidifying deposition are generally low in the western Continental U.S. (CONUS) but can be 13 higher in the eastern CONUS (ISA Appendix 7, Section 8.5.1). In the eastern CONUS, acid-14 sensitive ecosystems are generally located in upland, mountainous terrain underlain by 15 weathering resistant bedrock. Surface waters most sensitive to acidification are largely found in 16 the Northeast, southern Appalachian Mountains, Florida, the Upper Midwest, and the 17 mountainous West. (ISA, Appendix 8, section 8.5.1).

18 Acidification of freshwater ecosystems occurs in response to either N or S deposition 19 alone or in combination. This is because both N and S deposition can act as acidifying agents. 20 The effects of acidifying deposition on biogeochemical processes in soils have ramifications for 21 the water chemistry and biological functioning of associated surface waters. Surface water 22 chemistry integrates direct air-to-water deposition with deposition impacts on soil chemistry of hydrologically connected terrestrial ecosystems within the watershed (ISA, Appendices 4, 7 and 23 24 8). Acid-sensitive freshwater systems can either be chronically acidified or subject to occasional 25 episodes of decreased pH, decreased ANC, and increased inorganic Al concentration (ISA, 26 Appendix 7, section 7.1). 27 In this assessment, the impact of N and/or S deposition on aquatic acidification was 28 evaluated using a critical load (CL) approach. This CL approach provides a means of gauging

whether a group of lakes, streams, and rivers (i.e., waterbodies) in each area receives a level of N and/or S deposition that corresponds to that associated with a specified value for the water quality metric used as indicator of acidification. For this analysis, ANC was used as the indicator, with target levels identified to correspond to different levels of acidification-related

33 risk to biota. Depending on the ANC target, low CL values (i.e., less than 50 meq/m<sup>2</sup>/yr) may

34 mean that the watershed has a limited ability to neutralize the addition of acidic anions, and

5A-2

hence, it is susceptible to acidification. The greater the CL value, the greater the ability of the
 watershed to neutralize the additional acidic anions.

3 5A.1.1 Analysis Scales

4 A multi-scale analysis was completed that assessed aquatic acidification at three levels of 5 spatial extent: national, ecoregion, and case study (Figure 5A-1). For this analysis, the national-6 scale assessment included the CONUS only since there is insufficient data available for Hawaii, 7 Alaska, and the territories. The Omernik ecoregion classifications were used for the ecoregion-8 scale analyses. Case studies were selected for areas which were likely to be most impacted and 9 for which sufficient data was available. Further discussion of these spatial scales can be found 10 below. Since acidification of waterbodies is controlled by local factors such as geology, 11 hydrology, etc. the aquatic CLs for acidification are unique to the waterbody itself and 12 information about the waterbody, like water quality, is needed to determine its critical load. For 13 these reasons, CLs were determined at the waterbody level and then summarized at the national, 14 ecoregion, and case study level. The national assessment is a combined summary of aquatic 15 CLs across the CONUS.





1 It is important to note that aquatic ecosystems across the CONUS exhibit a wide range of 2 sensitivity to acidification because of a host of landscape factors, such as geology, hydrology, 3 soils, catchment scale, and vegetation characteristics that control whether a waterbody will be 4 impacted by acidifying deposition. Consequently, variations in ecosystem sensitivity must be 5 taken into account in order to characterize sensitive populations of waterbodies and relevant 6 regions across the CONUS. The EPA's Omernik Ecoregions classifications was used to define 7 ecologically relevant, spatial aggregated, acid sensitive regions across the CONUS in order to 8 better characterize the regional differences in the impact of deposition driven acidification 9 (Figure 5A-2). There are 25 Ecoregion II categories in the CONUS, each of which are further

10 subdivided into a total of 84 Level III categories in the CONUS.





12 Figure 5A-2. Omernik Ecoregion II areas with ecoregion III subdivisions

Ecoregions are areas of similarity regarding patterns in vegetation, aquatic, and terrestrial ecosystem components. Available ecoregion categorization schemes include the EPA's Omernik

- 15 classifications (Omernik, 1987). Omernik's ecoregions are categorized using a holistic, "weight-
- 16 of-evidence" approach in which the relative importance of factors may vary from region to

2 regions through the analysis of the patterns and the composition of biotic and abiotic phenomena 3 that affect or reflect differences in ecosystem quality and integrity. Factors include geology, 4 physiography, vegetation, climate, soils, land use, wildlife, and hydrology. Three hierarchical 5 levels were developed to distinguish coarser (more general) and finer (more detailed) 6 categorization. Level I is the coarsest level, dividing the CONUS into 12 ecoregions. At level II, 7 the continent is subdivided into 25 ecoregions. Level III is a further subdivision of level II and 8 divides North America into 105 ecoregions with 84 in the CONUS. Level IV is a subdivision of 9 level III into 967 ecoregions for the CONUS. 10 The case study scale represents the smallest scale at which we performed our analyses

region. The method used to map ecoregions is described in Omernik (1987) and classifies

and is intended to give some insight into the local impact of aquatic acidification. Five case
study areas across the U.S. were examined. These areas were the Shenandoah National Park,
White Mountain National Forest, Voyagers National Park, Sierra National Forest, and Rocky
Mountain National Park. These parks and national forest vary in their sensitivity to acidification,
but represent high value or protected ecosystems, such as Class 1 areas, wilderness, and national
forests.

17

1

#### 5A.1.2 Method - Aquatic Critical Load Approach

18 The impacts of N and/or S deposition on aquatic ecosystems were evaluated using a CL 19 approach. The CL approach was used to characterize the risk of N and/or S deposition on aquatic 20 acidification across the CONUS with a focus on acid sensitive areas. In this assessment, the CL 21 approach provides a means of gauging whether an individual or group of waterbodies (i.e., lake 22 or stream) in a given area receives an amount of deposition that results in the waterbody not 23 being able to achieve the target ANC level (as described in 5A.1.3). Critical load exceedances 24 were summarized at the national, ecoregion III, and case study levels to understand the spatial 25 extent of deposition driven acidication across the CONUS. Special consideration was given to 26 naturally occuring aquatic acidification in order to focus the analysis on deposition driven 27 impacts to aquatic biota. Uncertainty within the CL was also estimated and factored in the CL 28 exceedance determination.

29

#### 5A.1.3 Ecological Risk and Response

Risk in aquatic systems is estimated based on the acidification indicator, ANC, and
changes in this water quality metric related to N and/or S deposition. The evidence relates ANC
and other water quality indicators of acidification to biological and ecological effects (ISA
Appendix 8.3). The connection between SO<sub>2</sub> and NO<sub>X</sub> emissions, deposition of N and/or S, and
the acidification of surface waters is well documented in the eastern U.S. (ISA, Appendix
7;Driscoll et al., 2016).

1 The biological impact of acidifying deposition is mediated through changes in water 2 quality that in turn impact biota. Deposition of N and/or S can effect biogeochemical changes 3 that may induce biologically harmful effects. Surface water chemistry is then a good indicator of 4 the effects of acidification on the biotic integrity of freshwater ecosystems, because it integrates 5 soil and water processes that occur within a watershed. Changes in surface water chemistry 6 reflect the influence of acidic inputs from precipitation, gases, and particles, as well as local geology and soil conditions. Surface water chemical factors such as pH, Ca<sup>2+</sup>, ANC, base 7 8 cations, ionic metals concentrations, and DOC are affected by acid deposition and can affect the 9 structure and function of biological communities in lakes and streams (ISA Appendix 8.3).

10 The most widely used measure of surface water acidification, and subsequent recovery 11 under reduced acid deposition, is ANC. Inorganic Al and pH are also affected by acidic 12 deposition. All three water quality parameters are indicators of aquatic acidification for which 13 there is evidence of effects on aquatic systems including physiological impairment, reduced 14 fitness or death, alteration of species richness, community composition and structure, and 15 biodiversity in freshwater ecosystems. The evidence of effects on biota from aquatic 16 acidification indicates a range of severity with varying levels of ANC, pH and inorganic Al, with 17 effects on biota ranging from phytoplankton and invertebrates to fish communities (ISA, 18 Appendix 8, section 8.5).

19 As summarized in section 4.2.1.1.2 above, the evidence of effects on biota from aquatic 20 acidification indicates a range of severity with varying ANC levels. The evidence relates to biota 21 ranging from phytoplankton and invertebrates to fish communities. For example, a review by 22 Lacoul et al. (2011) of aquatic acidification effects on aquatic organisms in Atlantic Canada 23 observed that the greatest differences in phytoplankton species richness occurred across a pH 24 range of 4.7 to 5.5 (ANC range of 0 to 20  $\mu$ eq/L), just below the range (pH 5.5 to 6.5) where 25 bicarbonate becomes rapidly depleted in the water (ISA, Appendix 8, section 8.3.1.1). Under 26 acidifying conditions, these phytoplankton communities shifted from dominance by 27 chrysophytes, other flagellates, and diatoms to dominance by larger dinoflagellates. In benthic 28 invertebrates residing in sediments of acidic streams, Al concentration is a key influence on the 29 presence of sensitive species. Studies of macroinvertebrate species have reported reduced species 30 richness at lower pH, with the most sensitive group, mayflies, absent at the lowest levels. Values 31 of pH below 5 (which may correspond to approximant ANC concentrations below 0  $\mu$ eq/L)<sup>1</sup> were 32 associated with the virtual elimination of all acid sensitive mayfly and stonefly species over the 33 period from 1937-42 to 1984-85 in two streams in Ontario (Baker and Christensen, 1991). In a

<sup>&</sup>lt;sup>1</sup> pH and ANC were related to one another using a generalized relationship base on equilibrium with atmospheric CO<sub>2</sub> concentration (Cole and Prairie, 2010)

- 1 more recent study, Baldigo, et al., (2009) showed macroinvertebrate assemblages in the
- 2 southwestern Adirondack Mountains were severely impacted at pH <5.1, moderately impacted at
- 3 pH from 5.1 to 5.7, slightly impacted at pH from 5.7 to 6.4 and usually unaffected above pH 6.4
- 4 (Figure A5-3). In Atlantic Canada, Lacoul et al. (2011) found the median pH for sensitive
- 5 invertebrate species occurrence was between 5.2 and 6.1 (ANC of 10 and 80 µeq/L), below
- 6 which such species tended to be absent. For example, some benthic macroinvertebrates,
- 7 including several species of mayfly and some gastropods are intolerant of acid conditions and
- 8 only occur at pH  $\geq$ 5.5 (ANC 20 µeq/L) and  $\geq$ 6, (ANC 50 µeq/L) respectively. (ISA, Section
- 9 8.3.3).



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Figure 5A-3. Total macroinvertebrate species richness as a function of pH in 36 streams in western Adirondack Mountains of New York, 2003-2005. From Baldigo et al. (2009); see ISA, Appendix 8, section 8.3.3, and p. 8-12.

Responses among fish species and life stages to changes in ANC, pH and Al in surface
waters are variable. Early life stages such as larvae and smolts are more sensitive to acidic
conditions than the young-of-the-year, yearlings, and adults (Baker, et al., 1990; Johnson et al.,
1987; Baker and Schofield 1985). Studies showed a loss of fish whole-body sodium when stream

- 18 pH drops below 5.1 (ANC 0  $\mu$ eq/L) indicating that trout lost the ability to ionoregulate. Some
- 19 species and life stages experienced significant mortality in bioassays at relatively high pH ((e.g.,
- 20 pH 6.0–6.5; ANC 50-100  $\mu$ eq/L for eggs and fry of striped bass and fathead minnow)
- 21 (McCormick et al., 1989; Buckler et al., 1987)), whereas other species were able to survive at
- 22 quite low pH without adverse effects. Many minnows and dace (Cyprinidae) are highly sensitive

- 1 to acidity, but some common game species such as brook trout, largemouth bass, and
- 2 smallmouth bass are less sensitive (threshold effects at pH <5.0 to near 5.5; ANC 20 and 50
- $3 \mu eq/L$ ). A study by Neff et al. (2008), investigated the effects of two acid runoff episodes in the
- 4 Great Smoke Mountains National Park on native brook trout using an in-situ bioassay. The
- 5 resulting whole-body sodium concentrations before and after the episodes showed negative
- 6 impacts on physiology. More specifically, the reduction in whole-body sodium when stream pH
- 7 dropped below 5.1 (ANC 0 µeq/L) indicated that the trout had lost the ability to ionoregulate
- 8 (ISA, Appendix 8, section 8.3.6.1). See Figure A5-4 for fish species sensitivity based on
- 9 observations from field studies with supporting bioassays.



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**Figure 5A-4. Critical aquatic pH range for fish species.** Notes: Baker and Christensen (1991) generally defined bioassay thresholds as statistically significant increases in mortality or by survival rates less than 50% of survival rates in control waters. For field surveys, values reported represent pH levels consistently associated with population absence or loss. Source: Fenn et al. (2011) based on Baker and Christensen (1991) (ISA, Appendix 8, Figure 8-3).

As noted in the ISA, "[a]cross the eastern U.S., brook trout are often selected as a biological indicator of aquatic acidification because they are native to many eastern surface waters and because residents place substantial recreational and aesthetic value on this species" (ISA, Appendix 8, p. 8-26). Compared to other fish species in Appalachian streams, this species is relatively pH sensitive. For example, "[in many Appalachian mountain streams that have been acidified by acidic deposition, brook trout is the last fish species to disappear; it is generally lost at pH near 5.0 (MacAvoy and Bulger, 1995), which usually corresponds in these streams with

8 ANC near 0  $\mu$ eq/L (Sullivan et al., 2003)" (ISA, Appendix 8, p. 8-21).

9 As described in section 4.2.1 above episodic acidification during storm events can pose 10 risks in low ANC streams. For example, streams with ANC around 20 µeq/L or less at base flow 11 may be considered vulnerable to episodic acidification events that could reduce pH and ANC to 12 levels potentially harmful to brook trout and other species. Streams with suitable habitat and 13 annual average ANC greater than about 50 µeq/L are often considered suitable for brook trout in 14 southeastern U.S. streams and reproducing brook trout populations are expected (Bulger et al., 15 2000). Streams of this type "provide sufficient buffering capacity to prevent acidification from 16 eliminating this species and there is reduced likelihood of lethal storm-induced acidic episodes" 17 (ISA, Appendix 8, p. 8-26). Results of a study by Andren and Rydin (2012) suggested a 18 threshold less than 20 ug/L Al and pH higher than 5.0 for healthy brown trout populations by 19 exposing yearling trout to a pH and inorganic Al gradient in humic streams in Scandinavia (ISA, 20 Appendix 8, section 8.3.6.2). Another recently available study that investigated the effects of 21 episodic pH shifts fluctuations in waterbodies of eastern Maine reported that episodes resulting 22 in pH dropping below 5.9 (ANC of  $\sim$ 50 µeq/L) have the potential for harmful physiological 23 effects to Atlantic salmon smolts if coinciding with the smolt migration in eastern Maine rivers 24 (Liebich et al., 2011; ISA, Appendix 8, section 8.3.6.2).

25 There is often a positive relationship between pH or ANC and number of fish species, at 26 least for pH values between about 5.0 and 6.5, or ANC values between about 0 and 50 to 100 27 µeq/L (Cosby et al., 2006; Sullivan et al., 2006; Bulger et al., 1999). This is because energy cost 28 in maintaining physiological homeostasis, growth, and reproduction is high at low ANC levels 29 (Sullivan et al., 2003; Wedemeyer et al., 1990). As noted in section 4.2.1.1.2 above, surveys in 30 the heavily impacted Adirondack mountains found that lakes and streams having an annual 31 average ANC  $< 0 \mu eq/L$  and pH near or below 5.0 generally support few or no fish species to no 32 fish at all, as illustrated in Figure 5-3 below (Sullivan et al., 2006; ISA, Appendix 8, section 33 8.3.6.3.



Figure 5A-5. Number of fish species per lake *versus* acidity status, expressed as ANC, for
 Adirondack lakes. Notes: The data are presented as the mean (filled circles) of
 species richness within 10 µeq/L ANC categories, based on data collected by the
 Adirondacks Lakes Survey Corporation. Source: Modified from Sullivan et al.
 (2006). (ISA, Appendix 8, Figure 8-4)

7 The data presented in Figure 5A-5 above suggest that there could be a loss of fish species with

8 decreases in ANC below a threshold of approximately 50 to 100 µeq/L for lakes (Sullivan et al.,

9 2006). For streams in Shenandoah National Park, a statistically robust relationship between ANC

10 and fish species richness was also documented by Bulger et al., (2000). However, interpretation

11 of species richness relationship with ANC can be difficult and misleading, because more species

12 tend to occur in larger lakes and streams as compared with smaller ones, irrespective of acidity

13 (Sullivan et al., 2006) because of increased aquatic habitat complexity in larger lakes and streams

14 (Sullivan et al., 2003; ISA, Appendix 8, section 8.3.6.3).

15 The key biological/ecological effects on aquatic organisms that have been observed in 16 field and laboratory studies of different acidification levels, as described above, are summarized 17 below in the context of ANC ranges: <0, 0-20, 20-50, 50-80, and >80 µeq/L:

At ANC levels <0 µeq/L, aquatic ecosystems have exhibited low to a near loss of aquatic diversity and small population sizes. For example, planktonic and macroinvertebrates communities shift to the most acid tolerant species (Lacoul et al., 2011) and mayflies can be eliminated (Baker and Christensen, 1991). A near to complete loss of fish populations can occur, including non-acid sensitive native species such as brook trout (Salvelinus fontinalis), northern pike (Esox lucius), and others (Sullivan et al., 2003, 2006; Bulger et al., 2000), which is in most cases attributed to elevated inorganic monomeric Al</li>

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- concentration (Baldigo and Murdoch 1997). At this level, aquatic diversity is at its lowest
   (Bulger et al. 2000, Baldigo et al. 2009, Sullivan et al. 2006) with only acidophilic
   species being present.
- 4 In waterbodies with ANC levels between 0 and 20 µeg/L, acidophilic species dominate 5 other species (Matuszek and Beggs, 1988; Driscoll et al., 2001) and diversity is low 6 (Bulger et al. 2000, Baldigo et al. 2009, Sullivan et al. 2006). Plankton and 7 macroinvertebrate populations have been observed to decline, and acid-tolerant species 8 have outnumbered non-acid sensitive species (Liebich et al., 2011). Sensitive species are 9 often absent (e.g., brown trout, common shiner, etc.) while non-sensitive fish species populations may be reduced (Bulger et al. 2000). Episodic acidification events (e.g., 10 inflow with ANC <0  $\mu$ eq/L and pH< 5), may have lethal impacts on sensitive lifestages 11 12 of some biota, including brook trout and other fish species (Matuszek and Beggs, 1988; 13 Driscoll et al., 2001).
- Levels of ANC between 20 and 50 µeg/L have been associated with the loss and/or 14 15 reduction in fitness of aquatic biota that are sensitive to acidification in some 16 waterbodies. Such effects included reduced aquatic diversity (Kretser et al., 1989, Lawrence et al., 2015; Dennis, 1995) with many species missing such as Atlantic salmon 17 18 (Salmo salar) smolts, blacknose shiner (Notropis heterolepis) (Bulger et al., 2000, 19 Sullivan et al., 2006, Liebich et al., 2011). Comparatively, acid tolerant species, such as 20 brook trout may have moderate to healthy populations, (Kretser et al., 1989, Lawrence et 21 al., 2015; Dennis, 1995).
- 22 • At an ANC between 50 to 80 µeq L-1, the fitness and population size of only sensitive 23 species have been impacted. Levels of ANC above 50 µeg/L are considered suitable for 24 brook trout and most fish species because buffering capacity is sufficient to prevent the 25 likelihood of lethal episodic acidification events (Driscoll et al. 2001; Baker and 26 Christensen 1991). However, depending on other factors, the most sensitive species have 27 been reported to experience a reduction in fitness and/or population size in some waterbodies (e.g., Atlantic salmon smolts, blacknose shiner [Baldigo et al., 2009; Kretser 28 29 et al., 1989, Lawrence et al., 2015; Dennis, 1995]). Reduced fish species richness has also 30 been reported to be affected (Bulger et al., 2000 and Sullivan et al., 2006).
- Values of ANC >80 μeq/L have not generally been associated with harmful effects on
   biota (Bulger et al., 1999; Driscoll et al., 2001; Kretser et al., 1989; Sullivan et al., 2006).
- 33

#### 5A.1.4 Chemical Criterion and Critical Threshold

- 34 Most aquatic CL studies conducted in the U.S. use surface water ANC as the principal
- 35 metric of water quality change in response to changes in a N and/or S deposition, which is
- 36 known as the chemical criterion. The ANC is generally a more stable measurement than pH
- 37 because ANC is insensitive to changes in CO<sub>2</sub> and it reflects sensitivity and effects of
- 38 acidification in a linear fashion across the full range of ANC values. The critical threshold is then
- 39 the value of the chemical criterion (ANC) beyond which it is negatively impacted. For these
- 40 analyses, CLs were evaluated with respect to three different ANC thresholds: 20 µeq/L (minimal
- 41 protection), 30 (intermediate protection) and 50 µeq/L (moderate protection) that represent

1 specified harmful ecological effects based on results from Section 5A.1.3. Most aquatic CL

- 2 studies conducted in the U.S. since 2010 use an ANC of 20 and/or 50 µeq/L, because 20 µeq/L
- 3 provides protection for "natural" or "historical" range of ANC and 50 µeq/L provides overall
- 4 ecosystem protection (DuPont et al., 2005, McDonnell et. al. 2012, 2014, Sullivan et al., 2012a,
- 5 2012b, Lynch et al. 2022, Fakhraei et al. 2014, Lawrence et al., 2015). In the Mountain west,
- 6 vulnerable lakes and streams to deposition driven aquatic acidification are often found in the
- 7 mountains where surface water ANC levels are low and typically vary between 0 and 30 µeq/L
- 8 (Williams and Labou 2017, Shaw et al 2014). For these reasons, previous studies, and the
- 9 National Critical Loads Database (NCLD), used ANC threshold of 50 µeq/L for the eastern and
- 10 20  $\mu$ eq/L for the western CONUS (denoted as "50/20"  $\mu$ eq/L). An ANC of 80  $\mu$ eq/L was
- 11 considered; however, it was determined that many waterbodies, particularly, in acid sensitive
- 12 regions of CONUS never had an ANC that high and would never reach an ANC that high
- 13 naturally.
- 14

#### 5A.1.4.1 Natural Acidic Waterbodies

15 Some waterbodies are naturally acidic because of a host of factors, but most commonly 16 due to acidic rock within the waterbodies watershed, low base cation weathering rates linked to 17 the type of bedrock, and high DOC with the surface waters. Natural or historical level of ANC 18 concentration are typically above 20  $\mu$ eq/L (Sullivan et. el., 2012; Shaw et al 2014). Sullivan et 19 el., (2012) using MAGIC model simulations for pre-industrial (1850), suggested that there were 20 no acidic (ANC  $\leq 0 \mu eq/L$ ) and only ~6% of modeled lakes had ANC  $\leq 20 \mu eq/L$  in the 21 Adirondack mountains, NY. For these reasons, most recent CL studies (since 2010) use 20 22 µeq/L as a minimum ANC threshold. For waterbodies where their natural or historical level of 23 ANC is lower than the selected ANC threshold, the calculated CL is invalid or not achievable at any level of deposition. In nearly all known cases, historical ANC levels are above 20 µeq/L, 24 25 but not all waterbodies at the higher ANC levels of 50 µeq/L are able to reach this level. In those

cases, the CL was evaluated, but was not included in the results and summary assessments.

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#### 5A.1.5 Critical Load Data

- 28 Aquatic CLs used in this assessment came from the National Critical Load Database
- version 3.2.1 (Lynch et al., 2022), from studies identified in the ISA (e.g., Lawrence et al., 2015;
- 30 Fakhraei et al., 2014; Sullivan et al., 2012; Fakhraei et al., 2016). The NCLD is comprised of
- 31 CLs calculated from a host of common models: (1) steady-state mass-balance models such as the
- 32 Steady-State Water Chemistry (SSWC), (2) dynamic models such as Model of Acidification of
- 33 Groundwater In Catchment (MAGIC) (Cosby et al., 1985) or Photosynthesis EvapoTranspiration
- 34 Biogeochemical model (PnET-BGC) (Zhou et. al., 2011) run out to year 2100 or 3000 to model
- 35 steady-state conditions and (3) regional regression models that use results from dynamic models

- 1 to extrapolate to other waterbodies (McDonnell et. al., 2012 and Sullivan et al. 2012). These
- 2 approaches differ in the way watershed base cation weathering was determined (e.g., F-Factor or
- 3 dynamic model). Figure 5A-6 shows the unique locations for 13,000+ CLs used in this
- 4 assessment. Critical load waterbodies are concentrated in areas that are acid sensitive in the
- 5 eastern U.S. and the Rocky Mountain and Pacific Northwest regions of the west. Not all
- 6 waterbodies are sensitive to acidification. Small to medium size lakes size (>200 Ha) and
- 7 streams (1- 3 orders) tend to be the waterbodies that are impacted by deposition driven
- 8 acidification. Rivers are not typically impacted by deposition driven acidification. Data in the
- 9 NCLD is focused on waterbodies that are typically impacted by deposition driven acidification.
- 10 A waterbody is represented as a single CL value. In many cases, a waterbody has more than one
- 11 CL value calculated for it because different studies determined a value for the same waterbody.
- 12 When more than one CL exists, the CL from the most recent study was selected or averaged
- 13 when the publications are from the same timeframe.
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Figure 5A-6. Unique waterbody locations with critical loads used in this assessment. Lower critical load values are red and orange.
1	5A.1.5.1 Steady-State Water Chemistry (SSWC) model and F-Factor
2	Critical loads were derived from present-day water chemistry and are based on the
3	principle that excess base cation production within a catchment area should be equal to or greater
4	than the acid anion input, thereby maintaining the ANC above a pre-selected level (Scheffe et al.,
5	2014; Miller, 2011; Dupont et al., 2005; and Vermont Department of Environmental
6	Conservation (VDEC), 2003, 2004, 2012). This model assumes a mass balance and that all SO <sub>4</sub> <sup>2-</sup>
7	in runoff originates from sea salt spray and anthropogenic deposition. In the Steady State Water
8	Chemistry (SSWC) model, CL of acidity, CL(A), is calculated based on the principle that the
9	acid load should not exceed the non-marine, base cation inputs minus a nutrient base cation
10	uptake and ANC buffer to protect selected biota from being damaged (Eq. 5A-2):
11 12	$CL(A) = BC^{*}_{dep} + BC_{w} + -Bc_{u} - nANC_{crit} $ (5A-2)
13	Where:
14 15	BC <sup>*</sup> <sub>dep</sub> (BC; Ca+Mg+K+Na) = the sea-salt corrected non-anthropogenic deposition of base cations;
16	$BC_w$ (BC; Ca+Mg+K+Na) = the average watershed weathering flux;
17	Bc <sub>u</sub> (Bc: Ca+Mg+K) = the net long-term average uptake of base cations in the biomass
18	(i.e., the annual average removal of base cations due to harvesting);
19	$nANC_{crit}$ = the lowest ANC-flux that protects the biological communities. Bc <sub>u</sub> for these
20	studies was set to zero.
21	For these CLs, the SSWC model was modified to incorporate a simplified N framework
22	whereby N components that account for nitrogen removal from long-term nitrogen
23	immobilization and denitrification were included in the model (Eq. 5A-3):
24	$CL(A) = BC^{*}_{dep} + BC_{w} + N_{u} + N_{i} + N_{de} - Bc_{u} - nANC_{crit} $ (5A-3)
25	Where:
26	$N_u = N$ removal through removal of trees with harvesting;
27	$N_i = N$ removal from long-term N immobilization;
28	$N_{de} = N$ removal from the soil through microbial denitrification.
29 30 31 32	N <sub>i</sub> was set equal to 4.30 meq/m <sup>2</sup> -yr (McNulty et al., 2007) and N <sub>de</sub> was set equal to 7.14 meq/m <sup>2</sup> - yr (Ashby, et al., 1998) for sites in the east. For western states, a combined value of N <sub>i</sub> +N <sub>de</sub> = 11.0 eq/ha-yr was used based on Nanus et al., 2012. N <sub>u</sub> value varies depending on CL project. See below section "Critical Load Exceedance" regarding how exceedance of Critical
33	Loads of S, N and Combined S and N Deposition are calculated. In addition, exceedance for

these CLs can be determined using the N<sub>le</sub> (Henriksen and Posch, 2001) after removing the N
terms from (Eq. 5A-4):

3

$$Ex(A) = S_{dep} + N_{le} - CL(A)$$
(5A-4)

4 Where:

5  $N_{le}$  = the sum of the measured concentrations of nitrate (NO<sub>3</sub><sup>-</sup> eq/L) and ammonia (NH<sub>4</sub><sup>+</sup> 6 eq/L) in the runoff (Q<sub>s</sub> m/yr) as ([NO<sub>3</sub><sup>-</sup>]+[ NH<sub>4</sub><sup>+</sup>])\*Q<sub>s</sub>.

Equation 5A-4 determines the CL exceedance based on S deposition while incorporating
the present day measured (or assumed future) extent of N leaching.

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#### 5A.1.5.2 MAGIC Model and Regional Linear Regression Models for Estimating BC<sub>w</sub> Input to SSWC

For Sullivan et al., (2012) and McDonnell et al., (2012), CLs were derived using a modified form of the SSWC model (see Eq. 5A-3)<sup>-</sup> Additionally, base cation weathering was derived using a new method based on MAGIC model estimates of BC<sub>w</sub> and regional linear regression models (see Sullivan et al., 2012 and McDonnell et al., 2012), rather than the F-factor method for estimating BC<sub>w</sub>.

16 The MAGIC model was used to calculate watershed-specific BC<sub>w</sub> for input to regional 17 linear regression models that estimated BC<sub>w</sub> in watersheds without MAGIC values. The BCw 18 estimates were then used as input to the SSWC model. MAGIC is a lumped-parameter model of 19 intermediate complexity, developed to predict the long-term effects of acidic deposition on 20 surface water chemistry (Cosby et al., 1985). The model simulates soil solution chemistry, 21 weathering rates, and surface water chemistry to predict the monthly and annual average 22 concentrations of the major ions in these waters (see Cosby et al., 1985 for more details about the 23 model itself). The base cation weathering terms in MAGIC represent the catchment-average 24 weathering rates for the soil compartments. In a one soil-layer application of MAGIC, the 25 weathering rates in MAGIC thus reflect the catchment-average net supply of base cations to the 26 surface waters draining the catchment. The sum of the MAGIC weathering rates for the 27 individual base cations is therefore identical in concept to the base cation weathering term, BC<sub>w</sub>, 28 in the SSWC CL model (Eq. 5A-2). Base cation weathering rates in MAGIC are calibrated 29 parameters. The calibration procedure uses observed deposition of base cations, observed (or 30 estimated) base cation uptake in soils, observed stream water base cation concentrations, and 31 runoff (Qs). These observed input and output data provide upper and lower limits for internal 32 sources of base cations in the catchment soils. The two most important internal sources of base 33 cations in catchment soils are modeled explicitly by MAGIC: primarily mineral weathering and 34 soil cation exchange. During the calibration process, observed soil base saturation for each base 35 cation and observed soil chemical characteristics are combined with the observed input and

output data to partition the inferred net internal sources of base cations between weathering and
 base cation exchange.

3 Sullivan et al., (2012) and McDonnell et al., (2012) used the watershed-specific BC<sub>w</sub> to 4 develop a regional regression model for calculating watershed specific  $BC_w$  for 500+ monitoring 5 locations in the Appalachian Mountains of Virginia and West Virginia. Water chemistry and 6 landscape variables were used as the predictor variables in regression analyses to extrapolate 7 BC<sub>w</sub>. Each of the calibrated MAGIC study watersheds was placed in an Ecoregion category 8 based on which Ecoregion contained the maximum watershed area and three separate regression 9 models were developed for each Ecoregion (Table 5A-1). Watershed averages were used to 10 represent the spatial variability within each watershed for the landscape characteristics, except 11 for watershed area. Regression models were established using stepwise linear regression using 12 'best subsets' to evaluate candidate models and constrain the number of independent predictor 13 variables during model selection. Water quality predictor data was collected during several 14 regional surveys, as compiled by Sullivan and Cosby, 2004). One water quality sample, 15 generally collected during the spring between 1985 and 2001, was used to characterize each 16 watershed. Water quality data were derived from several regional surveys, including the National 17 Stream Survey (NSS), Environmental Monitoring and Assessment Program (EMAP), Virginia 18 Trout Stream Sensitivity Study (VTSSS), and stream surveys conducted in Monongahela 19 National Forest.

## Table 5A-1. Multiple Regression Equations to Estimate BC<sub>w</sub> from Either Water Chemistry and Landscape Variables or from Landscape Variables Alone, Stratified by Ecoregion. (McDonnell et al. 2012).

Ecoregion	n	Equation <sup>1</sup>	r <sup>2</sup>			
Central Appalachian	24	BC <sub>w</sub> = -37.5 + 0.6 (SBC*) + 0.9 (NO <sub>3</sub> ) + 0.006 (WS Area)	0.93			
Ridge and Valley	42	BC <sub>w</sub> = 107.0 + 0.5 (SBC*) - 0.06 (Elevation) - 3.2 (Slope)	0.86			
Blue Ridge	26	BC <sub>w</sub> = 27.1 + 0.6 (CALK*) + 0.6 (NO <sub>3</sub> )	0.90			
*SBC is the sum of base cations: CALK is calculated ANC						

23 24

### 5A.1.5.3 MAGIC model and Hurdle Modeling for Estimating BC<sub>w</sub> Input to SSWC

For McDonnell et al., 2014) and Povak et al., 2014), CLS were derived using a modified form of the SSWC model that excluded the N terms. Building on the framework of Sullivan et

27 al., (2012) and McDonnell et al., (2012), McDonnell et al. (2014) and Povak et al. (2014)

28 expanded the study area and developed new statistical models to better predict BC<sub>w</sub> and evaluate

29 CLs of S. Their study expanded the area to include the full Southern Appalachian Mountain

30 (SAM) region and surrounding terrain from northern Georgia to southern Pennsylvania, and

1 from eastern Kentucky and Tennessee to central Virginia and western North Carolina. As with 2 Sullivan et al., (2012) and McDonnell et al., (2012), the MAGIC model was used to calculate 3 watershed-specific BC<sub>w</sub> for 140 stream locations containing both measured soil chemistry and 4 water chemistry data (see section above for a description of MAGIC). In addition, McDonnell 5 et al., (2014) aggregated all known water quality data that totaled 933 sample locations in order 6 to develop a statistical model to predict ANC and BC<sub>w</sub> for all streams in the SAM region. Water chemistry data were collected between 1986 and 2009, with stream ANC calculated as the 7 equivalent sum of the base cation concentrations ( $Ca^{2+}$ ,  $Mg^{2+}$ ,  $K^+$ ,  $Na^+$ , ammonium [ $NH_4^+$ ]) 8 9 minus the sum of the mineral acid anion concentrations (chloride [Cl\_],  $NO_3^-$ ,  $SO_4^{2-}$ ). BC<sub>w</sub> was 10 estimated as the net internal source of base cations between weathering and base cation exchange 11 for the watershed based on the MAGIC model calibrations, which used observed stream, soil, 12 and atmospheric deposition data to match current observed stream and soil chemistry conditions. 13 With the use of a random forest regression modeling technique, a continuous  $BC_w$  layer was 14 regionalized using a suite of initial candidate predictor variables chosen to represent potential 15 broad- to fine-scale climatic, lithologic, topoedaphic, vegetative, and S deposition variables that have the potential to influence ANC and BC<sub>w</sub>. To represent the landscape conditions that 16 17 influence specific locations along a stream, all candidate landscape predictor variables were 18 expressed on a 30 m grid basis across the SAM's domain and were upsloped averaged based on 19 the technique described in McDonnell et al., (2012). This resolution allowed for the creation of 20 "flowpaths" for the development of a topographically determined stream network. This 21 approach allowed for a total of 140,504 watersheds which were represented (i.e., delineated) 22 with the use of a hydrologically conditioned based on digital elevation models (DEM). CLs were 23 then calculated with SSWC (Henriksen and Posch, 2001) with estimates of BCdep,  $BC_w$ ,  $Bc_u$ ,  $Q_s$ 24 and an ANC chemical criteria set to an value of 50 µeq/L for each stream node. See McDonnell 25 et al., (2014) and Povak et al., (2014) for additional methods detail.

26

#### 5A.1.6 Critical Load Exceedance

A critical load exceedance (Ex) is when deposition is greater than the critical load.
Critical Load exceedances define when the ecological resources are likely to be harmed by
deposition.

If N and/or S deposition is less than the aquatic CL, adverse ecological effects (e.g., reduced reproductive success, stunted growth, loss of biological diversity) are not anticipated, and recovery is expected over time if an ecosystem has been damaged by past exposure. When pollutant exposure is higher than, or "exceeds," the CL and the ecosystem continues to be exposed to damaging levels of pollutants. Critical loads and deposition estimates are uncertain and to have confidence in the exceedance it is important that this uncertainty is factored into the 1 calculation. Based on the CL uncertainty analysis (see section 5A-2), on average the magnitude

- 2 of the uncertainty for aquatic CLs is  $4.29 \text{ meq S/m}^2/\text{yr}$  or 0.69 Kg S/ha-yr and a confidence
- 3 interval of  $\pm 2.15 \text{ meq/m}^2/\text{yr}$  or  $\pm 0.35 \text{ Kg S/ha/yr}$ . For simplicity reasons, a 6.25 meq S/m<sup>2</sup>/yr or
- 4 1 Kg S/ha/yr range of uncertainty was used in the exceedance calculation. Within this range, it is
- 5 unclear whether the CL is exceeded. For that reason, CLs are exceeded where deposition is
- 6 above 3.125 meq S/m<sup>2</sup>/yr or 0.5 Kg S/ha/yr and are not exceeded where deposition is below
- 7  $3.125 \text{ meq S/m}^2/\text{yr or } 0.5 \text{ Kg S/ha/yr}$ . The exceedances that fall within this range are described

8 as being "at" the CL. A detailed discussion of exceedances can be found in Chapter VII:

9 Exceedance calculation of the 2015 ICP Modelling and Mapping Manual (see

10 <u>http://icpmapping.org/Latest\_update\_Mapping\_Manual).</u>

11 Aquatic CL exceedances can be considered with respect to S and combined N and

12 S deposition. When considering only S deposition (i.e., N deposition is zero), the exceedance is

13 expressed as the difference between the CL of S, total S deposition, and an uncertainty of  $\pm 3.125$ 

14 meq S/m<sup>2</sup>/yr or  $\pm 0.5$  Kg S/ha/yr (Eq. 5A-6).

15 
$$Exceedance (Ex) = (Total S deposition - CLS) > 3.125 meq S/m2-yr$$
 (5A-6)

In most cases, deposition of both S and N contributes to the exceedance. Calculating a combined S and N Ex is more complex because both S and N contribution to acidification needs to be factored in the exceedance. Given that not all N deposition to a watershed will contribute to acidification, the N deposition removed by long-term N processes in the soil and waterbody (e.g., N uptake and immobilization) defines a "minimum" CL for N. Nitrogen deposition inputs below what is removed do not acidify, but the amount above this minimum will likely contribute to acidification.

Exceedance of both N and S is a two-step calculation where if N removal is greater than N deposition, only S deposition contributes to the Ex (Eq. 5A-8). However, if deposition of N is greater than what is removed, the amount is not removed (Eq. 5A-9):

26 When minimum  $CLN \ge Total N$  deposition, then

27 
$$Ex(N+S) = Total S deposition - CLS$$
 (5A-8)

28 When minimum CLN < Total N deposition, then

29 
$$Ex(N+S) = Total S + N deposition - CLS + minimum CLN$$
 (5A-9)

30 There are different methods for determining the contribution of N deposition to aquatic 31 acidification. The section below described the two most common methods and how they are 32 handled in the CL exceedance calculations.

5A-18

#### 1 **5A.1.6.1 Deposition**

2 The amount of deposition used in the critical load exceedance calculation was determined 3 from the Total Deposition (TDep) model (https://nadp.slh.wisc.edu/committees/tdep/) (Schwede 4 and Lear (2014). See section 2.5.1 for more details. Both total N and S deposition were 5 determined to be the deposition level at the grid cell of the stream reach or lake location. For 6 each waterbody total N and S deposition was determined for each year from 2000 to 2020. 7 Three-year averages were calculated for these periods: 2000-03, 2007-09, 2014-16 and 2018-20 8 to be used in the different analyses. Critical load exceedances were then calculated for each of 9 these four periods and summed nationally and by Ecoregion III.

10

#### 5A.1.6.2 Acidifying Contribution of Nitrogen Deposition

11 Unlike sulfur, not all N deposition leads to acidification. In fact, in some systems, none 12 of the entering N deposition acidifies because it is retained in biomass (terrestrial and aquatic) 13 and soils or is lost to the atmosphere by denitrification. Determining the contributions of N 14 deposition that acidifies is difficult to estimate and uncertain because of the underlining 15 processes that store and release N in a watershed is complex, making them hard to measure or 16 model. Different methods have been developed to determine the amount of N deposition that 17 acidifies related to aquatic CL exceedances. There are two common approaches used in CL 18 studies: the first approach is based on the amount of "N leaching" to the waterbody determined 19 by the amount of dissolved N in the water measured as the concentration of nitrite and runoff as 20 presented in Henriksen and Posch, (2001) the second approach is the use of a "set value" based 21 on long-term estimate of N immobilization and denitrification as described by McNulty et al 22 (2007).

23 While the majority of atmospherically deposited N is either denitrified or accumulates in 24 watershed soils, vegetation, or groundwater (Galloway et al., 2008), the relative partitioning of N 25 loss via denitrification versus watershed storage is poorly known (Galloway et al., 2004). The 26 amount of N leaching to a waterbody that is not retained within the waterbody's biota is the 27 actual amount that contributes to acidification in the surface water. This depends on the amount 28 of N immobilized in the watershed, the amount exported to the drainage waters from the soils, 29 and how much uptake there is within the waterbody itself (Bergström 2010; ISA Appendix 30 9.1.1.2). As the different forms of N deposition enter a watershed, they undergo many 31 biogeochemical changes that result in N being stored in the soil and vegetation and being 32 released to the drainage water. As N deposition enters the watershed it can be quickly taken up 33 by the microorganisms in the soils and vegetation (particularly NH<sub>3</sub>) and incorporated into 34 biomass. This is the amount of N immobilized in the watershed. Nitrogen immobilization or 35 accumulation is the conversion of inorganic N to organic N. The amount that is immobilized can

5A-19

1 be variable. Lovett et al., (2000) found immobilization of N to be 49% to 90% of the

- 2 atmospheric input based on N measured in stream water. The variation is because of a host of
- 3 factors such as vegetation type, age of vegetation, soil type, soil condition, the amount of
- 4 nitrification, management activities, etc that control the amount of N accumulating. Gregor et
- 5 al., (2004) reported values of nitrogen immobilization for forest soil plots ranging from 2 to 5 kg
- 6 N ha/yr in colder climates and up to 10 kg N ha-yr in warmer climates.

Nitrogen is removed or exported from the watershed by being volatilized in fires,
denitrified or leached to drainage waters. Denitrification is the process by which nitrate is

- 9 converted into gaseous N, most commonly in water saturated soil, and returned to the
- 10 atmosphere. Like with immobilization, a host of factors control the rate of denitrification,
- 11 making it difficult to estimate at a site by site bases without directly measuring it. Dutch and
- 12 Ineson, (1990) ranged from 0.1–3.0 kg N/ha/yr while in well drained soils denitrification was
- 13 below 0.5 kg N/ha/yr, which is similar to Groffman et al., (2009) found denitrification in
- 14 temperate ecosystems had a mean value of 1.9 kg N ha/yr for forest soils. The remaining amount
- 15 of N that isn't volatilized, denitrified, or immobilized is leached in drainage water as nitrate or
- 16 dissolved organic nitrogen (DON) and has the potential to acidify surface waters. Nitrate
- 17 concentrations or DON in streams impacted by acidification (typically 1-3 order streams) is often
- 18 very low, near zero, during the growing season (Campbell et al., 2000; Perakis and Hedin 2002;
- 19 MacDonald et al., 2002 ; De Vries et al., 2007; Dise et al., 2009). This is because nearly all the
- 20 N entering the watershed is incorporated in the soil or vegetation.
- 21Recent studies from some regions of the U.S. (e.g., Eshleman et al. (2013); Driscoll et al.22(2016); Strock et al. (2014); Eshleman and Sabo (2016); ISA Appendix 7.1.5.1) showed declines23in concentrations of  $NO_3^-$  in surface waters that are consistent with declines in N deposition.
- 24 Using the Lake Multi-Scaled Geospatial and Temporal Database of the Northeast Lakes of the
- 25 U.S. (LAGOS-NE) containing water quality data from 2,913 lakes, Oliver et al. (2017) identified
- 26 atmospheric deposition as the main driver of declines in total N (TN) deposition and lake
- 27 TN:total P (TP) ratios from 1990 to 2011. In additional, monitored lakes and streams as part of
- 28 the EPA's Long-term Monitoring (LTM) program have average annual nitrate concentrations of
- 29 9.5 and 7.64  $\mu$ eq/L, respectively, from 1990 to 2018 (Table 5A-2). Average annual nitrate
- 30 concentrations have decreased during the past decade to 7.19 and 4.40  $\mu$ eq/L. These areas
- 31 receive 5 to 8 kg N/ha/yr deposition annually.
- 32 Recent studies from some regions of the U.S. (e.g., Eshleman et al. (2013); Driscoll et al.
- 33 (2016); Strock et al. (2014); Eshleman and Sabo (2016); ISA Appendix 7.1.5.1) showed declines
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- 1 atmospheric deposition as the main driver of declines in total N (TN) deposition and lake
- 2 TN:total P (TP) ratios from 1990 to 2011. In additional, monitored lakes and streams as part of
- 3 the EPA's Long-term Monitoring (LTM) program have average annual nitrate concentrations of
- 4 9.5 and 7.64 μeq/L, respectively, from 1990 to 2018 (Table 5A-3). Average annual nitrate
- 5 concentrations have decreased during the past decade to 7.19 and 4.40  $\mu$ eq/L. These areas
- 6 receive 5 to 8 kg N/ha/yr deposition annually.

		Average (95% CI)
Areas	Years	(µeq/L)
New England Lakes	1990 – 2018	2.36 (2.155 – 2.565)
	1990 – 1999	2.33 (1.947 – 2.713
	2000 – 2009	2.45 (2.165 – 2.745)
	2010 – 2018	0.56 (0.46 – 0.66)
Adirondacks Lakes	1990 – 2018	16.64 (15.966 – 17.318)
	1990 – 1999	18.48 (17.183 – 19.779)
	2000 – 2009	16.70 (15.602 – 17.796)
	2010 – 2018	13.82 (12.736 – 14.907)
Appalachian Streams	1990 – 2018	7.64 (7.092 – 8.187)
	1990 – 1999	11.50 (10.334 – 12.675)
	2000 - 2009	6.59 (5.774 – 7.40)
	2010 – 2018	4.40 (3.744 - 5.049)

1	Table 5A-2.	Average annual nitrate concentrations for the EPA's Long-term Monitoring
2		(LTM) program for lakes and streams.

3 We recognize that estimating the contribution of N to acidification of surface waters is 4 difficult and uncertain because N cycling in an ecosystem is inherently variable and data are 5 limited across the U.S. to model it, however, it is important to the review that an estimate be 6 determined for aquatic acidification. Given the availability of data and what was used in the 7 2008 review, we chose the N leaching method that uses water quality and runoff data to estimate 8 the amount of N deposition leaching to the drainage water that acidify (Henriksen and Posch 9 2001). 10 This method is based on Henriksen and Posch, (2001) where the exceedance for these 11 CLs is determined using the Nle (see Eq. 5A-10): Ex(N+S) = Total S deposition + Nle - CLS12 (5A-10)13 Where:

14Nle = the sum of the measured concentrations of nitrate (NO3<sup>-</sup>  $\mu$ eq/L) and ammonia15(NH4<sup>+</sup>  $\mu$ eq/L) in the runoff (Qs m/yr) as ([NO3<sup>-</sup>]+[ NH4<sup>+</sup>])\*Qs.

16 Factoring in the CL uncertainty Eq. 5A-11 is:

17 
$$Ex(N+S) = ((Total \ S \ deposition + Nle) - CLS) > 3.125 \ meq \ S/m^2 - yr$$
(5A-11)

18 The advantage of using a leaching estimate (Nle) is that for some waterbodies it is based 19 on a measured water quality value that integrates all the N processes occurring in the watershed. 20 However, it's a measurement of current conditions. Steady-state CLs are intended to represent 21 the long-term leaching amount, which may or may not be well represented under current

5A-22

2 able to immobilize most of the N deposition. However, that would not be the case for old growth 3 forests, which leach N at a much higher rate than younger forests (Vitousek and Reomers 1975, 4 Goodale et al. 2000), although this model has been questioned in recent years (Lovett et al. 5 2018). Old growth forests are thought of as the steady-state condition. If future forests are older, 6 then the leaching estimate based on current water quality would under-estimate the acidification 7 affect. But if future forests are like today's forests, then the leaching value would better 8 represent acidification impacts. 9 The Nle metric for calculating the contribution to acidification from N deposition is

conditions. For example, if a forest is a watershed is young, it would be growing fast, and be

10 based on the calculated flux of N to the waterbody estimated as the concentration of nitrate as N 11 within the waterbody and the annual surface water runoff to the waterbody. Actual measured N 12 leaching values are not typically collected across the U.S. For that reason, the only way to 13 estimate an annual leaching is to calculate it as a function of annual runoff (eq 5A-10), which we 14 recognize adds additional uncertainty into this estimated value. We chose to use an annual 15 runoff based on 30-year Normals that is included as a catchment parameter in NHD+2 16 (https://www.epa.gov/waterdata/nhdplus-national-hydrography-dataset-plus). Site-specific 17 catchment annual runoff values were used for each waterbody with a CL. We decided to use 18 NHD+2 annual runoff value because it would better reflect long-term and temporal patterns in 19 runoff that reflects the mass-balance steady-state CL approach. In addition, the same runoff 20 value was used for the CL which provided consistency across both CL parameters. 21 Measurement of nitrate linked to the CL value in the NCLDv3.2 can date back to the 22 1980s and for that reason do not reflect the current N leaching rates. Also, many of the 23 waterbodies with CLs have no Nitrate water quality measurements, hence, no way to calculate 24 the leaching directly. Another limitation is that the water quality measurement is from a single 25 sample and may not reflect the variability of nitrate during the year and for that reason may over 26 or under-estimate it's contribution. For waterbodies with no or limited Nitrate water quality 27 measurements, a "regional approach" was used to estimate a value of leaching for that CL. We 28 recognize this regional approach provides additional uncertainty to the leaching estimate; 29 however, it at least provides an integrated regional average estimate that is based on numerous 30 available water quality data and long-term runoff data at the catchment level where the waterbody is located. We recognize that multiple water quality measurements over many years 31 32 for each waterbody and waterbody specific runoff or flow would be more desirable to estimate 33 the contribution of N deposition that acidifies, however, those data are not readily available. 34 The regional aggregation was done at the ecoregion III and II levels. Water quality data 35 came from the NCLD3.2 associated with the CLs and was supplemented with data from EPA's 36 LTM program (https://www.epa.gov/power-sector/long-term-monitoring-temporally-integrated-

1 monitoring-ecosystems, equaling 16,900+ measurements across the CONUS. We decide to focus 2 on the water quality data within the NCLD 3.2 because it represents the type of waterbodies (i.e., 3 small lakes/ponds, 1-3 order streams, etc.) that the CLs are based on. Measurements from within 4 each ecoregion III, II, and I were averaged to create a single aggregated value, which was used to 5 replace the measured value for the CL. The ecoregion average for level III was used unless there 6 were less than 30 water quality measurements, in which case the level II ecoregion average was 7 used, and if there were less than 30 measurements in level II, the level I ecoregion average was 8 used. The ecoregion average could over-estimate the amount of leaching for a given waterbody 9 because most have very little leaching and near-zero values. See Table 5A-3 for the number of 10 measured used in the aggregation and N leaching value.

### 11Table 5A-3.Regional aggregation for determine average N leaching for ecoregion II and12III. Water quality data based on National Critical Database v3.2

Name	Code	No.	Average N Leaching meq/m2/yr
Ecoregion III			
Northern Appalachian and Atlantic Maritime Highlands	5.3.1	3729	0.7
Blue Ridge	8.4.4	2703	1.7
Southern Rockies	6.2.14	444	1.2
Ridge and Valley	8.4.1	1719	3.0
Middle Rockies	6.2.10	552	1.3
Sierra Nevada	6.2.12	566	1.0
Northern Lakes and Forests	5.2.1	894	0.6
Acadian Plains and Hills	8.1.8	630	0.5
Piedmont	8.3.4	573	4.8
Northeastern Coastal Zone	8.1.7	526	1.8
Central Appalachians	8.4.2	495	3.0
Idaho Batholith	6.2.15	212	8.8
Cascades	6.2.7	229	1.4
Southeastern Plains	8.3.5	413	5.6
Northern Piedmont	8.3.1	265	16.1
Wasatch and Uinta Mountains	6.2.13	114	1.7
Atlantic Coastal Pine Barrens	8.5.4	263	5.2
North Central Appalachians	5.3.3	230	2.7
Northern Allegheny Plateau	8.1.3	224	3.3
North Cascades	6.2.5	169	1.0
South Central Plains	8.3.7	157	0.6
Southwestern Appalachians	8.4.9	127	2.4
Columbia Mountains/Northern Rockies	6.2.3	96	0.9
Southern Coastal Plain	8.5.3	149	1.6

News	Code	Na	Average N Leaching
Name	Code	NO.	meq/m2/yr
Middle Atlantic Coastal Plain	8.5.1	118	13.5
Coast Range	7.1.8	119	4.0
Eastern Great Lakes Lowlands	8.1.1	92	1.5
Klamath Mountains	6.2.11	85	1.2
North Central Hardwood Forests	8.1.4	101	1.7
Interior Plateau	8.3.3	89	7.2
Blue Mountains	6.2.9	65	0.3
Ozark Highlands	8.4.5	61	4.0
Eastern Cascades Slopes and Foothills	6.2.8	32	0.6
Ouachita Mountains	8.4.8	51	3.2
Strait of Georgia/Puget Lowland	7.1.7	39	4.5
Mississippi Valley Loess Plains	8.3.6	41	0.4
Arkansas Valley	8.4.7	39	1.7
Arizona/New Mexico Mountains	13.1.1	27	2.2
California Coastal Sage, Chaparral, and Oak Woodlands	11.1.1	25	0.7
Central Basin and Range	10.1.5	17	1.1
Western Allegheny Plateau	8.4.3	37	2.4
Northern Basin and Range	10.1.3	20	5.3
Southern Michigan/Northern Indiana Drift Plains	8.1.6	36	0.4
Canadian Rockies	6.2.4	32	1.4
Cross Timbers	9.4.5	31	0.8
Ecoregion II			
Atlantic Highlands	5.3	3960	0.85
Mixed Wood Plains	8.1	1639	1.51
Ozark/Ouachita-Appalachian Forests	8.4	5259	2.34
Southeastern USA Plains	8.3	1568	6.55
Mississippi Alluvial and Southeast USA Coastal Plains	8.5	551	5.85
Mixed Wood Shield	5.2	896	0.62
Temperate Prairies	9.2	51	1.57
Western Cordillera	6.2	2596	1.78
South Central Semi-Arid Prairies	9.4	48	0.65
Upper Gila Mountains	13.1	27	2.23
Mediterranean California	11.1	49	0.70
Marine West Coast Forest	7.1	182	4.72
Cold Deserts	10.1	46	2.83

#### 5A.1.7 Ecoregions Sensitivity to Acidification

Not all areas of the CONUS are sensitive to deposition driven acidification. The CONUS
areas known to be sensitive include the Northeast, Southeast, upper Midwest, Rocky Mountains,

- 1 Sierra Never Mountains, and the Pacific Northwest (Figure 5A-7). Mountain regions are most
- 2 susceptible to acidification, particularly, the Appalachian Mountains from Maine to Georgia. In
- 3 order to appropriately characterize the level of acidification in sensitive areas across the CONUS,
- 4 ecoregions were used as the unit of spatial aggregation. The EPA's Total alkalinity of Surface
- 5 Water GIS layer (Omernik and Powers 1983) and modern ANC water quality measurements
- 6 were used to define which ecoregions were considered acid sensitive. The EPA's Total
- 7 Alkalinity GIS layer was developed in 1980's using water quality data to define regions of acid
- 8 sensitivity (Figure 5A-7a) (Omernik and Powers 1983). Water quality ANC measurements were
- 9 collected from over 15,000 measurements from a host of water quality networks, programs,
- 10 groups across the CONUS for the period from 1990 to 2018 (Figure 5A-7b).

a. Total Alkalinity



Figure 5A-7. The EPA's Total Alkalinity regions (a) and ANC water quality measurements across the CONUS (b) in units of µeq/L. Red and orange colors (regions or points) are those that are most acid sensitive with lighter colors are those which are least sensitive. Data presented here was used to determine which Ecoregion IIIs are acid sensitive.

Water quality measurements and total alkalinity (Omernik and Powers 1983) were used
to classify CONUS ecoregion IIIs (e.g., 84) into four acid sensitive classes: (1) most acid
sensitive (<50 µeq/L), (2) acid sensitive (50-100 µeq/L), (3) moderately acid sensitive (100-200 µeq/L), and (4) low or no acid sensitivity (>200 µeq/L). The four categories are based on what

11 Omernik and Powers (1983) and Greaver et al. (2012) used in their assessment (Table 5A-4). A

- 1 total of 23 ecoregions III were acid sensitive and another 7 ecoregions were moderately acid
- 2 sensitive for a total of 30. Fifty-four ecoregions had low or no evidence of acid sensitivity
- 3 across the CONUS (Table 5A-5 and Figure 5A-8). The acid sensitive ecoregions generally are
- 4 areas with mountains, high elevation terrain or water bodies in northern latitudes (northern areas
- 5 of Minnesota, Wisconsin and Michigan; and New England). The northern, non-mountainous
- 6 regions share attributes (e.g., growing season, vegetation, soils and geology) similar to
- 7 mountainous regions and typically are located in rural areas, often in tracts of designated
- 8 wilderness, park and recreation areas. Of the 30 acid sensitive ecoregions, the following three
- 9 ecoregions are located on eastern coastal plain: (1) Middle Atlantic Coastal Plain (8.5.1), (2)
- 10 Southern Coastal Plains (8.5.3), and (3) Atlantic Coastal Pine Barrens (8.5.4). Waterbodies in
- 11 these ecoregions tend to have higher DOC values >10 mg/L, which is indicative of natural
- 12 acidity.

Acid Sensitive Category	Criteria
Most Acid Sensitive Ecoregions	>25 ANC values less than 100 $\mu$ eq/L, > 75 ANC values from 100-200 $\mu$ eq/L and have total alkalinity areas < 50 $\mu$ eq/L
Acid Sensitive Ecoregions	>10 ANC values less than 100 $\mu$ eq/L, > 40 ANC values from 100-200 $\mu$ eq/L and have total alkalinity areas < 100 $\mu$ eq/L
Moderately Sensitive Ecoregions	>5 ANC values less than 100 $\mu$ eq/L, > 20 ANC values from 100-200 $\mu$ eq/L and have total alkalinity areas < 200 $\mu$ eq/L
Low or Non-sensitive Ecoregions	<5 ANC values less than 100 $\mu$ eq/L, < 20 ANC values from 100-200 $\mu$ eq/L and have total alkalinity areas >200 $\mu$ eq/L

13 Table 5A-4. Acid sensitive Categories and criteria used to define each one.



Figure 5A-8. Ecoregion III grouped in three acid sensitivity classes. The dark colors indicate acid sensitive ecoregions. Points are ANC concentrations below 200 µeq/L.

5

1 2

3

Ecoregion III Code	No. Critical Loads	Total No. ANC Values	No. ANC values <200 µeq/L	Total Alkalinity Area (µeq/L)	Acid Sensitive Category
5.2.1	839	1074	933	50	Most Acid Sensitive Ecoregions
5.3.1	2851	2053	2203	50	Most Acid Sensitive Ecoregions
5.3.3	216	242	290	50	Most Acid Sensitive Ecoregions
6.2.10	496	324	190	50	Most Acid Sensitive Ecoregions
6.2.12	353	359	503	50	Most Acid Sensitive Ecoregions
6.2.14	372	327	163	50	Most Acid Sensitive Ecoregions
8.1.7	565	488	289	50	Most Acid Sensitive Ecoregions
8.1.8	494	492	513	50	Most Acid Sensitive Ecoregions
8.3.5	390	432	352	50	Most Acid Sensitive Ecoregions
8.4.1	1292	1394	1192	50	Most Acid Sensitive Ecoregions
8.4.2	372	420	511	50	Most Acid Sensitive Ecoregions
8.4.4	1972	1136	1535	50	Most Acid Sensitive Ecoregions
8.5.3	142	228	247	50	Most Acid Sensitive Ecoregions
8.5.4	234	130	178	50	Most Acid Sensitive Ecoregions
6.2.5	162	155	120	50	Most Acid Sensitive Ecoregions
8.3.4	508	455	112	100	Most Acid Sensitive Ecoregions
6.2.7	179	244	209	50	Most Acid Sensitive Ecoregions
8.1.3	199	223	55	100	Acid Sensitive Ecoregions
8.3.7	153	165	58	50	Acid Sensitive Ecoregions
8.5.1	105	183	51	50	Acid Sensitive Ecoregions
8.4.8	42	73	61	50	Acid Sensitive Ecoregions
8.4.9	117	64	51	50	Acid Sensitive Ecoregions
6.2.13	96	139	87	100	Acid Sensitive Ecoregions
6.2.15	188	164	155	50	Moderately Sensitive Ecoregions
8.1.4	94	162	33	200	Moderately Sensitive Ecoregions
6.2.9	63	91	21	100	Moderately Sensitive Ecoregions
6.2.3	86	147	44	50	Moderately Sensitive Ecoregions
8.4.7	31	59	34	100	Moderately Sensitive Ecoregions
6.2.11	81	105	16	100	Moderately Sensitive Ecoregions
8.4.6	23	31	23	100	Moderately Sensitive Ecoregions
6.2.4	31	42	8	100	Low or Non-sensitive Ecoregions
8.3.1	231	211	9	100	Low or Non-sensitive Ecoregions
7.1.8	115	154	17	200	Low or Non-sensitive Ecoregions
8.4.3	35	114	2	100	Low or Non-sensitive Ecoregions
8.3.3	71	114	2	200	Low or Non-sensitive Ecoregions
6.2.8	27	43	1	>200	Low or Non-sensitive Ecoregions
7.1.7	38	51	10	200	Low or Non-sensitive Ecoregions
8.1.1	83	97	3	>200	Low or Non-sensitive Ecoregions
8.3.6	41	61	15	200	Low or Non-sensitive Ecoregions

1 Table 5A-5. Ecoregion III results for acid sensitivity.

Ecoregion III Code	No. Critical Loads	Total No. ANC Values	No. ANC values <200 µeq/L	Total Alkalinity Area (µeq/L)	Acid Sensitive Category
13.1.1	25	64	3	>200	Low or Non-sensitive Ecoregions
8.3.2	18	115	10	100	Low or Non-sensitive Ecoregions
9.4.2	5	144	5	>200	Low or Non-sensitive Ecoregions
10.1.4	3	56	1	>200	Low or Non-sensitive Ecoregions
10.1.5	16	87	2	200	Low or Non-sensitive Ecoregions
5.2.2	2	26	2	>200	Low or Non-sensitive Ecoregions
10.1.3	20	80	4	>200	Low or Non-sensitive Ecoregions
8.1.5	15	80	2	>200	Low or Non-sensitive Ecoregions
8.2.1	10	38	2	>200	Low or Non-sensitive Ecoregions
9.6.1	0	7	2	>200	Low or Non-sensitive Ecoregions
8.1.6	33	131	0	>200	Low or Non-sensitive Ecoregions
8.4.5	56	111	0	>200	Low or Non-sensitive Ecoregions
9.4.5	26	96	0	>200	Low or Non-sensitive Ecoregions
9.2.3	26	180	0	>200	Low or Non-sensitive Ecoregions

#### 1 5A.2 ANALYSIS RESULTS

2 The aquatic acidification assessment is intended to estimate the ecological exposure and 3 risk posed to aquatic ecosystems from the acidification effects of S and/or N deposition to 4 sensitive regions across the CONUS. The CL itself indicates how sensitive the waterbody is to 5 inputs of acidic deposition of S and/or N. In Figure 5A-6, a CL indicates the amount of acidic 6 input of total S and/or N deposition that a waterbody can neutralize and still maintain an ANC of 7 50  $\mu$ eq/L. Watersheds with CL values less than 100 meq/m<sup>2</sup>-yr (red and orange circles) are most 8 sensitive to surface water acidification, whereas watersheds with values greater than 100 9 meq/m<sup>2</sup>/yr (yellow and green circles) are the least sensitive sites. Most sensitive waterbodies are 10 located along the Appalachian Mountains range, the upper Mid-west, and the Rocky Mountain 11 range in the west, which correspond to the same regions as the acid sensitive Ecoregion IIIs 12 (Figure 5A-7).

#### 13

#### 5A.2.1 Results of National Scale Assessment of Risk

14 A total of 13,824 unique waterbodies across the CONUS had calculated CLs. Table 5A-6

15 summarizes the percent of waterbodies with CLs that are less than 2, 6, 12, 18 kg S/ha,

16 indicating most CLs used in this analysis are less than 18 kg S/ha. Table 5A-7 contains a

17 summary of CL exceedances for S only and S and N combined for average deposition from

18 2018-20, 2014-16, 2010-12, 2006-08, and 2001-03 that is greater than the amount of deposition

19 the waterbodies could neutralize and still maintain the ANC thresholds for an ANC of 20, 30,

20 and 50 µeq/L.

1Table 5A-6.Percent of waterbodies with critical loads less than 2, 6, 12, and 18 Kg S/Ha2for critical loads based on an ANC limit of 20, 30, and 50 µeq/L

Critical Load Kg/Ha (meq/m²-yr)	Percent of Waterbodies Grouped by ANC threshold				
	20 µeq/L	30 µeq/L	50 µeq/L		
2 (12.5)	3%	5%	11%		
6 (37.5)	14%	17%	25%		
12 (75)	36%	39%	45%		
18 (112.5)	52%	55%	58%		

6

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Table 5A-7.Summary of national aquatic critical load exceedances by ANC thresholds<br/>and deposition periods. The percent of modeled waterbodies where<br/>deposition from 2018-20, 2014-16, 2010-12, 2006-08, and 2001-03 is above the<br/>critical load and error of 3.125 meq/m²-yr. "All Values" includes all critical<br/>loads. "CL>0 Values" includes only critical loads greater than 0.

ANC	Sulfur Only		Sulfur and Nitrogen		
Threshold	All Values	CL>0 Values Only	All Values	CL>0 Values Only	
	l	Deposition from 20	)18-20		
20	2%	1%	2%	2%	
30	3%	2%	4%	2%	
50	9%	4%	9%	5%	
50/20	7%	4%	8%	4%	
		Deposition from 20	)14-16		
20	3%	3%	3%	3%	
30	5%	4%	5%	4%	
50	11%	6%	12%	7%	
50/20	10%	6%	10%	7%	
		Deposition from 20	)10-12		
20	5%	5%	6%	5%	
30	8%	7%	9%	7%	
50	15%	11%	16%	11%	
50/20	14%	10%	15%	11%	
		Deposition from 20	06-08		
20	17%	16%	18%	17%	
30	21%	19%	21%	20%	
50	28%	24%	29%	25%	
50/20	27%	23%	28%	24%	
Deposition from 2001-03					
20	22%	22%	23%	23%	
30	26%	25%	27%	25%	
50	33%	28%	33%	29%	
50/20	31%	28%	32%	28%	

- 1 Table 5A-8 includes both numbers and percent exceedances for the CONUS for the four-2 deposition time periods and three ANC thresholds. Exceedance rates (e,g, percent of 3 waterbodies that exceed the CL) are the lowest for the least protective CL of 20 µeq/L and the 4 highest rate for most protective CL of 50 µeq/L. For the most recent deposition period of 2018-5 20, 2%, 3%, and 9% of the modeled waterbodies received levels of total S deposition that 6 exceeded their CL with CL thresholds of 20, 30, and 50 µeq/L, respectively. The percent of 7 exceeded waterbodies for combined total S and N are slightly higher than S only percents at 2%, 8 4%, and 9% of the modeled waterbodies for CL thresholds of 20, 30, and 50 µeq/L based on 9 short-term leaching of nitrate to the surface water. This indicates that most of the N deposition 10 entering the watershed is retained within the watershed and/or converted to gaseous N (e.g., N<sub>2</sub>O,
- $N_2$ , etc.). For all other deposition time periods, exceedance rates are only slightly higher (1-2%)
- 12 when considering both N and S deposition compared to just S deposition only.

## 13Table 5A-8.National aquatic critical load exceedances based on <u>all critical load</u> values by14ANC thresholds and deposition periods. The numbers and percent of15modeled waterbodies where deposition from 2018-20, 2014-16, 2010-12, 2006-1608, and 2001-03 is above the critical load and error of 3.125 meq/m²-yr.

		Sulfur Only		Sulfur Only Sulfur and Nitroge		
ANC						
Threshold	Class	No.	Percent	No.	Percent	
	De	eposition fr	om 2018-20			
20	>CL	234	2%	266	2%	
	<cl< td=""><td>13375</td><td>97%</td><td>13333</td><td>96%</td></cl<>	13375	97%	13333	96%	
	atCL	215	2%	225	2%	
30	>CL	452	3%	496	4%	
	<cl< td=""><td>13078</td><td>95%</td><td>13033</td><td>94%</td></cl<>	13078	95%	13033	94%	
	atCL	294	2%	295	2%	
50	>CL	1203	9%	1262	9%	
	<cl< td=""><td>12218</td><td>88%</td><td>12132</td><td>88%</td></cl<>	12218	88%	12132	88%	
	atCL	403	3%	430	3%	
5020	>CL	1023	7%	1075	8%	
	<cl< td=""><td>12416</td><td>90%</td><td>12344</td><td>89%</td></cl<>	12416	90%	12344	89%	
	atCL	385	3%	405	3%	
	De	eposition fr	om 2014-16			
20	>CL	423	3%	465	3%	
	<cl< td=""><td>13137</td><td>95%</td><td>13089</td><td>95%</td></cl<>	13137	95%	13089	95%	
	atCL	264	2%	270	2%	
30	>CL	680	5%	724	5%	
	<cl< td=""><td>12807</td><td>93%</td><td>11750</td><td>85%</td></cl<>	12807	93%	11750	85%	
	atCL	337	2%	1350	10%	

		Sulfur Only Sulfur and Nitroge		nd Nitrogen	
ANC					
Threshold	Class	No.	Percent	No.	Percent
50	>CL	1512	11%	1591	12%
	<cl< td=""><td>11859</td><td>86%</td><td>11750</td><td>85%</td></cl<>	11859	86%	11750	85%
	atCL	453	3%	483	3%
5020	>CL	423	3%	465	3%
	<cl< td=""><td>13137</td><td>95%</td><td>13089</td><td>95%</td></cl<>	13137	95%	13089	95%
	atCL	264	2%	270	3%
	D	eposition fr	om 2010-12	1	
20	>CL	748	5%	798	6%
	<cl< td=""><td>12731</td><td>92%</td><td>12670</td><td>92%</td></cl<>	12731	92%	12670	92%
	atCL	345	2%	356	3%
30	>CL	1122	8%	1192	9%
	<cl< td=""><td>12271</td><td>89%</td><td>12190</td><td>88%</td></cl<>	12271	89%	12190	88%
	atCL	431	3%	442	3%
50	>CL	2114	15%	2215	16%
	<cl< td=""><td>11206</td><td>81%</td><td>11099</td><td>80%</td></cl<>	11206	81%	11099	80%
	atCL	504	4%	510	4%
5020	>CL	1918	14%	2013	16%
	<cl< td=""><td>11424</td><td>83%</td><td>11324</td><td>80%</td></cl<>	11424	83%	11324	80%
	atCL	482	3%	487	4%
	D	eposition fr	om 2006-08	1	I
20	>CL	2328	17%	2433	18%
	<cl< td=""><td>10994</td><td>80%</td><td>10871</td><td>79%</td></cl<>	10994	80%	10871	79%
	atCL	502	4%	520	4%
30	>CL	2845	21%	2962	21%
	<cl< td=""><td>10450</td><td>76%</td><td>10322</td><td>75%</td></cl<>	10450	76%	10322	75%
	atCL	529	4%	540	4%
50	>CL	3911	28%	4035	29%
	<cl< td=""><td>9384</td><td>68%</td><td>9266</td><td>67%</td></cl<>	9384	68%	9266	67%
	atCL	529	4%	523	4%
5020	>CL	3710	2%	3825	28%
	< <u>C</u>	9609	70%	9492	69%
	atCl	505	4%	57	4%
	 	eposition fr	om 2006-08		.,.
20	>CI	3064	22%	3191	23%
20		10271	74%	10156	7.3%
 	atCl	489	4%	477	3%
30		35.97	26%	360/	27%
		079/	ZU /0	0683	Z1 /0 700/
		3104 AE2	20/	3003	20/
E O		403	3% 220/	447	3% 220/
50	>0L	4004	33%	4011	33%

		Sulfur Only		Sulfur and Nitrogen	
ANC Threshold	Class	No.	Percent	No.	Percent
	<cl< td=""><td>8905</td><td>64%</td><td>8807</td><td>64%</td></cl<>	8905	64%	8807	64%
	atCL	415	3%	406	3%
5020	>CL	4313	31%	4410	32%
	<cl< td=""><td>9124</td><td>66%</td><td>9030</td><td>65%</td></cl<>	9124	66%	9030	65%
	atCL	387	3%	384	3%

1 Table 5A-9 includes both numbers of waterbodies and percent exceedances for the 2 CONUS for the four-deposition time periods and four ANC thresholds where CLs less than or 3 equal to zero were removed from the exceedance percents. CLs less than or equal to zero are 4 very sensitive waterbodies that naturally could not meet the ANC threshold at any level of 5 deposition. The percent of modeled waterbodies with negative CLs is the lowest for an ANC 6 threshold of 20 µeq/L at 0.4% and the highest for an ANC threshold of 50 µeq/L at 4.6%. For 7 ANC thresholds of 30 and 50/20  $\mu$ eq/L (eastern U.S = 50  $\mu$ eq/L, western U.S. = 20  $\mu$ eq/L), the 8 exceedances were 1.3 and 3.5% (Figure 5A-9). Factoring in negative CLs, exceedances for the 9 most recent deposition period of 2018-2020, 1%, 2%, 4%, and 4% of the modeled waterbodies 10 received levels of total S deposition that exceeded their CL with CL thresholds of 20, 30, 50, and 11 50/20 µeq/L, respectively. The percent of exceeded waterbodies for combined total S and/or N 12 are slightly higher than S only percents at 2%, 2%, 5%, and 4% of the modeled waterbodies for 13 CL thresholds of 20, 30, 50, and  $50/20 \mu eq/L$  based on short-term leaching of nitrate to the 14 surface water. For the deposition period of 2001-2003, exceedance percentages for Sulfur only 15 were much higher at 22%, 25%, 29, and 29% for CL thresholds of 20, 30, 50, and 50/20 µeq/L.

# 16Table 5A-9.National aquatic critical load exceedances based on critical loads greater17than 0 by ANC thresholds and deposition periods. Zero or negative critical18loads were excluded from this summary. The numbers and percent of19modeled waterbodies where deposition from 2018-20, 2014-16, 2010-12, 2006-2008, and 2001-03 is above the critical load and error of 3.125 meq/m²-yr.

		Sulfur Only		Sulfur ar	nd Nitrogen
ANC	Class	Na	Demonst	Na	Demonst
Inresnoid	Class	NO.	Percent	NO.	Percent
		Deposition fro	om 2018-20		
20	>CL	182	1%	214	2%
	<cl< td=""><td>13375</td><td>97%</td><td>13333</td><td>97%</td></cl<>	13375	97%	13333	97%
	atCL	215	2%	225	2%
30	>CL	279	2%	323	2%
	<cl< td=""><td>13078</td><td>96%</td><td>13033</td><td>95%</td></cl<>	13078	96%	13033	95%
	atCL	293	2%	294	2%
50	>CL	566	4%	624	5%

		Sulfur Only		Sulfur and Nitroger	
ANC Threshold	Class	No.	Percent	No.	Percent
	<cl< td=""><td>12218</td><td>93%</td><td>12132</td><td>92%</td></cl<>	12218	93%	12132	92%
	atCL	401	3%	429	3%
5020	>CL	544	4%	596	4%
	<cl< td=""><td>12416</td><td>93%</td><td>12344</td><td>92%</td></cl<>	12416	93%	12344	92%
	atCL	385	3%	405	3%
I		Deposition fro	m 2014-16		
20	>CL	371	3%	413	3%
	<cl< td=""><td>13137</td><td>95%</td><td>13089</td><td>95%</td></cl<>	13137	95%	13089	95%
	atCL	264	2%	270	2%
30	>CL	506	4%	550	4%
	<cl< td=""><td>12807</td><td>94%</td><td>12730</td><td>93%</td></cl<>	12807	94%	12730	93%
	atCL	337	2%	370	3%
50	>CL	873	7%	952	7%
	<cl< td=""><td>11859</td><td>90%</td><td>11750</td><td>89%</td></cl<>	11859	90%	11750	89%
	atCL	453	3%	483	4%
5020	>CL	845	6%	921	7%
	<cl< td=""><td>12060</td><td>90%</td><td>11959</td><td>90%</td></cl<>	12060	90%	11959	90%
	atCL	440	3%	465	3%
		Deposition fro	m 2010-12	·	
20	>CL	696	5%	746	5%
	<cl< td=""><td>12731</td><td>92%</td><td>12670</td><td>92%</td></cl<>	12731	92%	12670	92%
	atCL	345	3%	356	3%
30	>CL	948	7%	1018	7%
	<cl< td=""><td>12271</td><td>90%</td><td>12190</td><td>89%</td></cl<>	12271	90%	12190	89%
	atCL	431	3%	442	3%
50	>CL	1475	11%	1576	12%
	<cl< td=""><td>11206</td><td>85%</td><td>11099</td><td>84%</td></cl<>	11206	85%	11099	84%
	atCL	504	4%	510	4%
5020	>CL	1439	11%	1534	11%
	<cl< td=""><td>11424</td><td>86%</td><td>11324</td><td>85%</td></cl<>	11424	86%	11324	85%
	atCL	482	4%	487	4%
		Deposition fro	m 2006-08		
20	>CL	2276	17%	2381	17%
	<cl< td=""><td>10994</td><td>80%</td><td>10871</td><td>79%</td></cl<>	10994	80%	10871	79%
	atCL	502	4%	520	4%
30	>CL	2671	20%	2788	20%
	<cl< td=""><td>10450</td><td>77%</td><td>10322</td><td>76%</td></cl<>	10450	77%	10322	76%
	atCL	529	4%	540	4%
50	>CL	3272	25%	3396	26%
	<cl< td=""><td>9384</td><td>71%</td><td>9266</td><td>70%</td></cl<>	9384	71%	9266	70%

		Sulf	ur Only	Sulfur and Nitrogen	
ANC Threshold	Class	No.	Percent	No.	Percent
	atCL	529	4%	523	4%
50/20	>CL	3231	24%	3346	25%
	<cl< td=""><td>9609</td><td>72%</td><td>9492</td><td>71%</td></cl<>	9609	72%	9492	71%
	atCL	505	4%	507	4%
		Deposition fro	om 2001-03		
20	>CL	3012	22%	3139	23%
	<cl< td=""><td>10271</td><td>75%</td><td>10156</td><td>74%</td></cl<>	10271	75%	10156	74%
	atCL	489	4%	477	3%
30	>CL	3413	25%	3520	26%
	<cl< td=""><td>9784</td><td>72%</td><td>9683</td><td>71%</td></cl<>	9784	72%	9683	71%
	atCL	453	3%	447	3%
50	>CL	3865	29%	3972	30%
	<cl< td=""><td>8905</td><td>68%</td><td>8807</td><td>67%</td></cl<>	8905	68%	8807	67%
	atCL	415	3%	406	3%
50/20	>CL	3834	29%	3931	29%
	<cl< td=""><td>9124</td><td>68%</td><td>9030</td><td>68%</td></cl<>	9124	68%	9030	68%
	atCL	387	3%	384	3%



Figure 5A-9. Critical load exceedance percentages by ANC thresholds and deposition years. Shade bars are the percent of waterbodies that have zero or negative critical loads and can't reach the given ANC threshold.

1 Figures 5A-10 to 5A-29 show mapped exceedances across the CONUS for S only for CL 2 thresholds of 20, 30, 50, and 50/20 µeq/L for positive CLs only. Figure 5A-30 highlights the 3 locations of waterbodies that have calculated negative CLs (grey dots). These are waterbodies 4 that are highly sensitive to acidification and likely naturally acidic. These waterbodies exceed the 5 calculated CL at any deposition amount. For these reasons, these sites have been removed from 6 the assessment. At their given ANC threshold, exceedance maps for S and/or N combined are not 7 included because they show the same pattern of exceedances and because exceedance rates are 8 only slightly higher for combined N and/or S deposition. Most exceedances occur in New 9 England, the Adirondacks, the Appalachian Mountain range (New England to Georgia), the 10 upper Midwest, Florida, and the Sierra Nevada mountains in California. Waterbodies in Florida 11 that exceed the CL are likely not related to deposition of S, but instead are related to high levels 12 of natural acidity in these drainage waters. These drainage waters tend to be naturally high in 13 dissolved organic carbon, causing these systems to be acidic. 14



Figure 5A-10. Critical load exceedance (Ex) for S only total deposition from 2001-03 for an ANC threshold of 20  $\mu$ eq/L. a. Blue dots are waterbodies with sulfur deposition below the CL and uncertainty (Ex < -3.125 meq/m<sup>2</sup>-yr). b. Red dots are waterbodies with sulfur deposition above the CL and uncertainty (Ex < 3.125 meq/m<sup>2</sup>-yr). Yellow dots are near the CL and based on the uncertainty cannot be determined if they exceed or not (-3.125 > Ex < 3.125 meq/m<sup>2</sup>-yr).



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Figure 5A-11. Critical load exceedance (Ex) for S only total deposition from 2001-03 for an ANC threshold of 30  $\mu$ eq/L. a. Blue dots are waterbodies with sulfur deposition below the CL and uncertainty (Ex < -3.125 meq/m<sup>2</sup>-yr). b. Red dots are waterbodies with sulfur deposition above the CL and uncertainty (Ex < 3.125 meq/m<sup>2</sup>-yr). Yellow dots are near the CL and based on the uncertainty cannot be determined if they exceed or not (-3.125 > Ex < 3.125 meq/m<sup>2</sup>-yr).



Figure 5A-12. Critical load exceedance (Ex) for S only total deposition from 2001-03 for an ANC threshold of 50  $\mu$ eq/L. a. Blue dots are waterbodies with sulfur deposition below the CL and uncertainty (Ex < -3.125 meq/m<sup>2</sup>-yr). b. Red dots are waterbodies with sulfur deposition above the CL and uncertainty (Ex < 3.125 meq/m<sup>2</sup>-yr). Yellow dots are near the CL and based on the uncertainty cannot be determined if they exceed or not (-3.125 > Ex < 3.125 meq/m<sup>2</sup>-yr).



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Figure 5A-13. Critical load exceedance (Ex) for S only total deposition from 2001-03 for an ANC threshold of 50 for the eastern and 20  $\mu$ eq/L for Western CONUS. a. Blue dots are waterbodies with sulfur deposition below the CL and uncertainty (Ex < -3.125 meq/m<sup>2</sup>-yr). b. Red dots are waterbodies with sulfur deposition above the CL and uncertainty (Ex < 3.125 meq/m<sup>2</sup>-yr). Yellow dots are near the CL and based on the uncertainty cannot be determined if they exceed or not (-3.125 > Ex < 3.125 meq/m<sup>2</sup>-yr).



Figure 5A-14. Critical load exceedance (Ex) for S only total deposition from 2006-08 for an ANC threshold of 20  $\mu$ eq/L. a. Blue dots are waterbodies with sulfur deposition below the CL and uncertainty (Ex < -3.125 meq/m<sup>2</sup>-yr). b. Red dots are waterbodies with sulfur deposition above the CL and uncertainty (Ex < 3.125 meq/m<sup>2</sup>-yr). Yellow dots are near the CL and based on the uncertainty cannot be determined if they exceed or not (-3.125 > Ex < 3.125 meq/m<sup>2</sup>-yr).



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Figure 5A-15. Critical load exceedance (Ex) for S only total deposition from 2006-08 for an ANC threshold of 30  $\mu$ eq/L. a. Blue dots are waterbodies with sulfur deposition below the CL and uncertainty (Ex < -3.125 meq/m<sup>2</sup>-yr). b. Red dots are waterbodies with sulfur deposition above the CL and uncertainty (Ex < 3.125 meq/m<sup>2</sup>-yr). Yellow dots are near the CL and based on the uncertainty cannot be determined if they exceed or not (-3.125 > Ex < 3.125 meq/m<sup>2</sup>-yr).



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Figure 5A-16. Critical load exceedance (Ex) for S only total deposition from 2006-08 for an ANC threshold of 50  $\mu$ eq/L. a. Blue dots are waterbodies with sulfur deposition below the CL and uncertainty (Ex < -3.125 meq/m<sup>2</sup>-yr). b. Red dots are waterbodies with sulfur deposition above the CL and uncertainty (Ex < 3.125 meq/m<sup>2</sup>-yr). Yellow dots are near the CL and based on the uncertainty cannot be determined if they exceed or not (-3.125 > Ex < 3.125 meq/m<sup>2</sup>-yr).



Figure 5A-17. Critical load exceedance (Ex) for S only total deposition from 2006-08 for an ANC threshold of 50 for the eastern and 20  $\mu$ eq/L for Western CONUS. a. Blue dots are waterbodies with sulfur deposition below the CL and uncertainty (Ex < -3.125 meq/m<sup>2</sup>-yr). b. Red dots are waterbodies with sulfur deposition above the CL and uncertainty (Ex < 3.125 meq/m<sup>2</sup>-yr). Yellow dots are near the CL and based on the uncertainty cannot be determined if they exceed or not (-3.125 > Ex < 3.125 meq/m<sup>2</sup>-yr).





Figure 5A-18. Critical load exceedance (Ex) for S only total deposition from 2010-12 for an ANC threshold of 20  $\mu$ eq/L. a. Blue dots are waterbodies with sulfur deposition below the CL and uncertainty (Ex < -3.125 meq/m<sup>2</sup>-yr). b. Red dots are waterbodies with sulfur deposition above the CL and uncertainty (Ex < 3.125 meq/m<sup>2</sup>-yr). Yellow dots are near the CL and based on the uncertainty cannot be determined if they exceed or not (-3.125 > Ex < 3.125 meq/m<sup>2</sup>-yr).



Figure 5A-19. Critical load exceedance (Ex) for S only total deposition from 2010-12 for an ANC threshold of 30  $\mu$ eq/L. a. Blue dots are waterbodies with sulfur deposition below the CL and uncertainty (Ex < -3.125 meq/m<sup>2</sup>-yr). b. Red dots are waterbodies with sulfur deposition above the CL and uncertainty (Ex < 3.125 meq/m<sup>2</sup>-yr). Yellow dots are near the CL and based on the uncertainty cannot be determined if they exceed or not (-3.125 > Ex < 3.125 meq/m<sup>2</sup>-yr).





Figure 5A-20. Critical load exceedance (Ex) for S only total deposition from 2010-12 for an ANC threshold of 50  $\mu$ eq/L. a. Blue dots are waterbodies with sulfur deposition below the CL and uncertainty (Ex < -3.125 meq/m<sup>2</sup>-yr). b. Red dots are waterbodies with sulfur deposition above the CL and uncertainty (Ex < 3.125 meq/m<sup>2</sup>-yr). Yellow dots are near the CL and based on the uncertainty cannot be determined if they exceed or not (-3.125 > Ex < 3.125 meq/m<sup>2</sup>-yr).


Figure 5A-21.	Critical load exceedance (Ex) for S only total deposition from 2010-12 for an ANC threshold of 50 for the eastern and 20 $\mu$ eq/L for Western CONUS a. Blue dots are waterbodies with sulfur deposition below the CL and uncertainty (Ex < -3.125 meq/m <sup>2</sup> -yr). b. Red dots are waterbodies with sulfur deposition above the CL and uncertainty (Ex < 3.125meq/m <sup>2</sup> -yr). Yellow dots are near the CL and based on the uncertainty cannot be determined if they exceed or not (-3.125 > Ex < 3.125 meq/m <sup>2</sup> -yr).
	determined if they exceed of not $(-5.125 \times Ex < 5.125 \text{ meq/m} - yr)$ .





Figure 5A-22. Critical load exceedance (Ex) for S only total deposition from 2014-16 for an ANC threshold of 20 μeq/L. a. Blue dots are waterbodies with sulfur deposition below the CL and uncertainty (Ex < -3.125 meq/m<sup>2</sup>-yr). b. Red dots are waterbodies with sulfur deposition above the CL and uncertainty (Ex < 3.125 meq/m<sup>2</sup>-yr). Yellow dots are near the CL and based on the uncertainty cannot be determined if they exceed or not (-3.125 > Ex < 3.125 meq/m<sup>2</sup>-yr).



Figure 5A-23. Critical load exceedance (Ex) for S only total deposition from 2014-16 for an ANC threshold of 30  $\mu$ eq/L. a. Blue dots are waterbodies with sulfur deposition below the CL and uncertainty (Ex < -3.125 meq/m<sup>2</sup>-yr). b. Red dots are waterbodies with sulfur deposition above the CL and uncertainty (Ex < 3.125 meq/m<sup>2</sup>-yr). Yellow dots are near the CL and based on the uncertainty cannot be determined if they exceed or not (-3.125 > Ex < 3.125 meq/m<sup>2</sup>-yr).

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Figure 5A-24. Critical load exceedance (Ex) for S only total deposition from 2014-16 for an ANC threshold of 50  $\mu$ eq/L. a. Blue dots are waterbodies with sulfur deposition below the CL and uncertainty (Ex < -3.125 meq/m<sup>2</sup>-yr). b. Red dots are waterbodies with sulfur deposition above the CL and uncertainty (Ex < 3.125 meq/m<sup>2</sup>-yr). Yellow dots are near the CL and based on the uncertainty cannot be determined if they exceed or not (-3.125 > Ex < 3.125 meq/m<sup>2</sup>-yr).



Figure 5A-25. Critical load exceedance (Ex) for S only total deposition from 2014-16 for an ANC threshold of 50 for the eastern and 20  $\mu$ eq/L for Western CONUS. a. Blue dots are waterbodies with sulfur deposition below the CL and uncertainty (Ex < -3.125 meq/m<sup>2</sup>-yr). b. Red dots are waterbodies with sulfur deposition above the CL and uncertainty (Ex < 3.125 meq/m<sup>2</sup>-yr). Yellow dots are near the CL and based on the uncertainty cannot be determined if they exceed or not (-3.125 > Ex < 3.125 meq/m<sup>2</sup>-yr).



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Figure 5A-26. Critical load exceedance (Ex) for S only total deposition from 2018-20 for an ANC threshold of 20  $\mu$ eq/L. a. Blue dots are waterbodies with sulfur deposition below the CL and uncertainty (Ex < -3.125 meq/m<sup>2</sup>-yr). b. Red dots are waterbodies with sulfur deposition above the CL and uncertainty (Ex < 3.125 meq/m<sup>2</sup>-yr). Yellow dots are near the CL and based on the uncertainty cannot be determined if they exceed or not (-3.125 > Ex < 3.125 meq/m<sup>2</sup>-yr).





Figure 5A-27. Critical load exceedance (Ex) for S only total deposition from 2018-20 for an ANC threshold of 30  $\mu$ eq/L. a. Blue dots are waterbodies with sulfur deposition below the CL and uncertainty (Ex < -3.125 meq/m<sup>2</sup>-yr). b. Red dots are waterbodies with sulfur deposition above the CL and uncertainty (Ex < 3.125 meq/m<sup>2</sup>-yr). Yellow dots are near the CL and based on the uncertainty cannot be determined if they exceed or not (-3.125 > Ex < 3.125 meq/m<sup>2</sup>-yr).



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Figure 5A-28. Critical load exceedance (Ex) for S only total deposition from 2018-20 for an ANC threshold of 50  $\mu$ eq/L. a. Blue dots are waterbodies with sulfur deposition below the CL and uncertainty (Ex < -3.125 meq/m<sup>2</sup>-yr). b. Red dots are waterbodies with sulfur deposition above the CL and uncertainty (Ex < 3.125 meq/m<sup>2</sup>-yr). Yellow dots are near the CL and based on the uncertainty cannot be determined if they exceed or not (-3.125 > Ex < 3.125 meq/m<sup>2</sup>-yr).



2 Figure 5A-29. Critical load exceedance (Ex) for S only total deposition from 2018-20 for 3 an ANC threshold of 50 for the eastern and 20 µeq/L for Western CONUS. 4 a. Blue dots are waterbodies with sulfur deposition below the CL and 5 uncertainty (Ex < -3.125 meq/m<sup>2</sup>-yr). b. Red dots are waterbodies with 6 sulfur deposition above the CL and uncertainty ( $Ex < 3.125 \text{ meq/m}^2$ -yr). Yellow dots are near the CL and based on the uncertainty cannot be 8 determined if they exceed or not  $(-3.125 > \text{Ex} < 3.125 \text{ meq}/\text{m}^2\text{-yr})$ . 9

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Figure 5A-30. Critical load exceedance (EX) for S only deposition from 2018-20 for an
ANC threshold: a. 20, b. 30, c. 50, d. 50/20 μeq/L for CONUS. Grey dots
are waterbodies where the critical load is zero or negative and was
excluded from the summary analysis. Red dots are waterbodies where
total sulfur is above the CL and uncertainty and yellow dots are where the
Ex is between -3.125 and 3.125 meq/m<sup>2</sup>-yr are near the CL and based on
the CL uncertainty cannot be determined if they exceed or not.

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## **5A.2.2 Ecoregion Analyses**

10 Acidification of waterbodies is controlled by local factors such as geology, hydrology,

11 etc. For this reason, aquatic CLs for acidification are unique to the waterbody itself and

12 information about the waterbody, like water quality, is needed to determine its CL.

13 Unfortunately, not all waterbodies within an ecoregion have sufficient data to calculate a CL.

14 This is the case for many ecoregion IIIs (from this point on ecoregion IIIs will be referred to as

15 ecoregions), except for ones that historically are known to be in acid sensitive areas since acid

16 sensitive areas typically have been heavily sampled, hence, contain many CLs (see Figure 5A-

17 31). These areas tend to be in the eastern CONUS in such ecoregions as Central Appalachians,

18 Northern Appalachian and Atlantic Maritime Highlands, and the Blue Ridge. Areas in the

- 1 Rockies and Sierra Nevada also have been sampled extensively and contain many CLs. More
- 2 CLs in an ecoregion helps to capture the spatial variability of acid sensitive areas across the
- 3 landscape and provide a more accurate measurement of the impact of deposition driven
- 4 acidification. Ecoregions with few CLs, however, fail to capture the spatial variability of acid
- 5 sensitive areas, which in turn reduces the accuracy of the percentile CL value and limits our
- 6 confidence in the estimated percent of exceedances. For this reason, ecoregions containing
- 7 greater than 50 CLs were the focus of this analysis while ones with less than 10 values were
- 8 included in the summary tables but excluded from the analysis.
- 9 For the CONUS there are a total of 84 ecoregions, 69 of which had at least one CL.
- 10 Eleven ecoregions had 9 or less CLs and 58 ecoregions had 10 or more. Of the 58 ecoregions,
- 11 however, only 32 had 50 or more CLs. The Northern Appalachian and Atlantic Maritime
- 12 Highlands ecoregion had the most CLs at 2,858 (see Table 5A-10).
- 13 The  $50^{\text{th}}$  to  $90^{\text{th}}$  ecoregion CLs varied greatly among ecoregions from 4.4 to 136.1
- 14 Kg/ha/yr (27.3 to 850.6 meq/m<sup>2</sup>/yr) for sulfur with an ANC threshold of 20  $\mu$ eq/L to 3.9 to 134.9
- 15 Kg/ha/yr (24.6 to 843.1 meq/m<sup>2</sup>/yr) for sulfur with an ANC threshold of 50  $\mu$ eq/L. Lower
- 16 percentile ecoregion values indicate higher sensitivity and risk for acidification. The most
- 17 sensitive ecoregions include Sierra Nevada, Southern Coastal Plain, Idaho Batholith, Atlantic
- 18 Coastal Pine Barrens, Blue Ridge, Middle Rockies, Wasatch and Uinta Mountains, Southern
- 19 Rockies, and Central Appalachian and Atlantic Maritime Highlands. See Tables 5A-10 and 5A-
- 20 11 for 70<sup>th</sup>, 90<sup>th</sup> Ecoregion CLs.



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2 Figure 5A-31. Locations of aquatic critical loads mapped across Ecoregions III.

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# Table 5A-10.Summary of Sulfur only critical loads by Ecoregions III by ANC thresholds<br/>of 20 and 30 µeq/L in Units = Kg S/ha-yr). Included ecoregions with more<br/>than 10 values.

Ecoregion III		ANC Threshold = 20			ANC Threshold = 30			
Name	Code	No.	70th	90th	Min.	70th	90th	Min.
Northern Appalachian and Atlantic Maritime Highlands	5.3.1	2851	9.7	4.8	0.0	8.8	4.0	0.0
Ridge and Valley	8.4.1	1292	11.6	5.9	0.1	10.8	5.2	0.2
Blue Ridge	8.4.4	1972	9.2	5.4	0.1	7.7	4.2	0.3
Northern Lakes and Forests	5.2.1	839	5.1	3.0	0.7	4.7	2.6	0.1
Northeastern Coastal Zone	8.1.7	565	16.4	8.1	1.3	15.3	7.2	0.0
Middle Rockies	6.2.10	496	9.2	5.2	0.5	8.4	4.5	0.2
Acadian Plains and Hills	8.1.8	494	11.4	5.5	0.2	10.6	4.9	0.0
Piedmont	8.3.4	508	16.0	8.7	0.9	15.0	7.7	0.1
Southern Rockies	6.2.14	372	7.4	3.9	0.3	6.5	3.2	0.0
Central Appalachians	8.4.2	372	8.5	5.3	0.3	7.6	4.2	0.2
Sierra Nevada	6.2.12	353	5.3	1.9	0.2	5.5	0.9	0.0
Southeastern Plains	8.3.5	390	14.2	4.9	0.4	13.7	4.6	0.1
Atlantic Coastal Pine Barrens	8.5.4	234	6.3	2.1	0.4	6.3	1.8	0.0
Northern Piedmont	8.3.1	231	40.0	16.8	1.5	39.2	15.8	1.0
North Central Appalachians	5.3.3	216	14.6	8.3	1.9	13.5	7.2	0.8
Northern Allegheny Plateau	8.1.3	199	22.2	11.8	0.2	21.2	10.9	1.1
Idaho Batholith	6.2.15	188	10.4	5.7	1.7	9.4	4.2	0.2
Cascades	6.2.7	179	14.4	4.3	0.0	15.2	5.8	0.2
North Cascades	6.2.5	162	26.3	12.4	5.2	24.8	10.1	1.5
Southern Coastal Plain	8.5.3	142	4.4	1.6	0.2	4.3	1.4	0.0
Coast Range	7.1.8	115	48.6	15.7	6.1	47.0	15.0	5.9
Middle Atlantic Coastal Plain	8.5.1	105	16.6	8.2	1.6	15.7	7.3	0.9
Wasatch and Uinta Mountains	6.2.13	96	11.0	7.7	2.1	10.4	6.7	1.6
North Central Hardwood Forests	8.1.4	94	23.0	5.8	2.8	22.0	4.8	0.1
Columbia Mountains/Northern Rockies	6.2.3	86	19.9	7.1	0.1	20.6	7.2	1.1
Eastern Great Lakes Lowlands	8.1.1	83	51.5	19.8	4.2	50.6	18.9	3.9
Klamath Mountains	6.2.11	81	27.6	12.4	7.3	26.5	11.6	6.2
Interior Plateau	8.3.3	71	66.8	10.9	5.3	65.8	9.8	3.2
Blue Mountains	6.2.9	63	18.1	8.6	3.6	16.7	7.5	2.5
South Central Plains	8.3.7	153	11.2	4.1	0.8	10.3	3.3	0.3
Ozark Highlands	8.4.5	56	48.3	13.5	2.8	47.4	12.5	1.7
Southwestern Appalachians	8.4.9	117	14.3	10.3	6.4	13.2	9.2	5.3
Ouachita Mountains	8.4.8	42	13.1	7.2	6.3	12.2	6.3	4.5
Strait of Georgia/Puget Lowland	7.1.7	38	28.9	10.5	4.6	28.3	9.2	3.6
Western Allegheny Plateau	8.4.3	35	18.7	8.2	5.0	17.7	7.0	4.3
Southern Michigan/Northern Indiana Drift Plains	8.1.6	33	11.5	5.8	2.1	10.8	4.4	1.3

Ecoregion III			ANC T	hresholo µeq/L	d = 20	ANC T	ANC Threshold = 30 µeq/L			
Name	Code	No.	70th	90th	Min.	70th	90th	Min.		
Arkansas Valley	8.4.7	31	14.9	6.3	3.4	14.1	5.4	2.6		
Canadian Rockies	6.2.4	31	41.8	8.3	3.5	40.4	7.8	1.6		
Western Corn Belt Plains	9.2.3	26	14.8	5.8	4.6	14.0	4.6	2.9		
Cross Timbers	9.4.5	26	11.3	7.1	2.9	10.0	5.2	0.8		
Eastern Cascades Slopes and Foothills	6.2.8	27	21.5	6.6	3.6	20.9	5.9	2.5		
Arizona/New Mexico Mountains	13.1.1	25	20.3	11.7	10.1	19.7	10.8	9.4		
Willamette Valley	7.1.9	24	65.2	25.5	8.3	63.2	24.9	7.4		
Boston Mountains	8.4.6	23	20.3	9.3	6.4	19.5	8.6	5.4		
Southern and Baja California Pine-Oak Mountains	11.1.3	22	25.2	3.4	1.1	27.4	5.5	2.1		
Central Irregular Plains	9.2.4	21	14.2	5.4	4.5	13.0	4.1	2.5		
California Coastal Sage, Chaparral, and Oak Woodlands	11.1.1	21	34.9	4.6	2.7	34.4	3.8	2.3		
Northern Basin and Range	10.1.3	20	19.1	10.1	3.2	18.7	8.9	1.7		
Mississippi Alluvial Plain	8.5.2	19	12.5	4.7	0.6	11.3	3.6	3.5		
Interior River Valleys and Hills	8.3.2	18	39.1	5.8	5.8	37.5	4.7	4.4		
Driftless Area	8.1.5	15	54.5	25.0	17.8	54.2	24.2	17.0		
Western Gulf Coastal Plain	9.5.1	16	52.2	20.6	10.3	51.6	19.7	9.4		
Central Basin and Range	10.1.5	16	45.4	21.5	8.7	44.4	20.3	6.4		
Eastern Corn Belt Plains	8.2.4	14	14.4	4.5	3.8	13.2	3.6	3.1		
Erie Drift Plain	8.1.10	14	18.6	5.8	4.1	17.6	4.8	2.8		
Mississippi Valley Loess Plains	8.3.6	41	14.5	4.2	1.6	13.4	3.1	0.5		
East Central Texas Plains	8.3.8	10	16.6	1.2	0.3	23.3	8.0	8.0		
Southeastern Wisconsin Till Plains	8.2.1	10	136.1	16.7	15.0	135.7	14.8	13.6		

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Table 5A-11. Summary of Sulfur only critical loads by Ecoregions III by ANC thresholds of 50 and 50/20 µeq/L in Units = Kg S/ha-yr). Included ecoregions with more than 10 values.

Ecoregion			ANC Threshold = 50			ANC Threshold =			
	1			µeq/L		50	/20 µeq/	L	
Name	Code	No.	70th	90th	Min.	70th	90th	Min.	
Northern Appalachian and Atlantic Maritime Highlands	5.3.1	2851	7.5	2.7	0.0	7.5	2.7	0.0	
Ridge and Valley	8.4.1	1292	9.2	3.8	0.0	9.2	3.8	0.0	
Blue Ridge	8.4.4	1972	5.3	2.0	0.0	5.3	2.0	0.0	
Northern Lakes and Forests	5.2.1	839	3.9	1.7	0.1	3.9	1.7	0.1	
Northeastern Coastal Zone	8.1.7	565	14.2	5.8	0.3	14.2	5.8	0.3	
Middle Rockies	6.2.10	496	6.8	3.2	0.0	9.2	5.2	0.5	
Acadian Plains and Hills	8.1.8	494	9.6	4.0	0.0	9.6	4.0	0.0	
Piedmont	8.3.4	508	13.2	5.7	0.2	13.2	5.7	0.2	
Southern Rockies	6.2.14	372	5.2	1.9	0.1	7.4	3.9	0.3	
Central Appalachians	8.4.2	372	6.1	2.7	0.1	6.1	2.7	0.1	
Sierra Nevada	6.2.12	353	7.5	1.8	0.0	5.3	1.9	0.2	
Southeastern Plains	8.3.5	390	12.6	3.9	0.1	12.6	3.9	0.1	
Atlantic Coastal Pine Barrens	8.5.4	234	5.9	1.6	0.1	5.9	1.6	0.1	
Northern Piedmont	8.3.1	231	38.2	14.4	1.3	38.2	14.4	1.3	
North Central Appalachians	5.3.3	216	11.9	5.5	0.4	11.9	5.5	0.4	
Northern Allegheny Plateau	8.1.3	199	19.6	9.9	0.5	19.6	9.9	0.5	
Idaho Batholith	6.2.15	188	7.8	1.7	0.2	10.4	5.7	1.7	
Cascades	6.2.7	179	15.8	5.0	0.1	14.4	4.3	0.0	
North Cascades	6.2.5	162	23.9	8.3	0.3	26.3	12.4	5.2	
Southern Coastal Plain	8.5.3	142	4.7	1.2	0.1	4.7	1.2	0.1	
Coast Range	7.1.8	115	42.5	14.0	4.9	48.6	15.7	6.1	
Middle Atlantic Coastal Plain	8.5.1	105	15.9	6.1	1.0	15.9	6.1	1.0	
Wasatch and Uinta Mountains	6.2.13	96	8.9	5.2	1.1	11.0	7.7	2.1	
North Central Hardwood Forests	8.1.4	94	20.7	3.7	0.0	20.7	3.7	0.0	
Columbia Mountains/Northern Rockies	6.2.3	86	20.6	6.0	0.2	19.9	7.1	0.1	
Eastern Great Lakes Lowlands	8.1.1	83	50.9	17.1	0.7	50.9	17.1	0.7	
Klamath Mountains	6.2.11	81	24.2	10.0	4.1	27.6	12.4	7.3	
Interior Plateau	8.3.3	71	69.4	11.8	0.9	69.4	11.8	0.9	
Blue Mountains	6.2.9	63	14.5	6.2	0.3	18.1	8.6	3.6	
South Central Plains	8.3.7	153	8.7	1.9	0.1	9.5	1.9	0.1	
Ozark Highlands	8.4.5	56	58.6	11.1	0.0	58.6	11.1	0.0	
Southwestern Appalachians	8.4.9	117	10.9	7.0	3.1	10.9	7.0	3.1	
Ouachita Mountains	8.4.8	42	10.2	4.8	0.4	10.2	4.8	0.4	
Strait of Georgia/Puget Lowland	7.1.7	38	28.9	8.0	1.6	28.9	10.5	4.6	
Western Allegheny Plateau	8.4.3	35	15.7	5.1	3.0	15.7	5.1	3.0	
Southern Michigan/Northern Indiana Drift Plains	8.1.6	33	9.5	3.8	0.3	9.5	3.8	0.3	

Ecoregion	Ecoregion			hresholo µeq/L	d = 50	ANC 50	ANC Threshold = 50/20 µeq/L		
Name	Code	No.	70th	90th	Min.	70th	90th	Min.	
Arkansas Valley	8.4.7	31	12.5	4.7	1.2	12.5	4.7	1.2	
Canadian Rockies	6.2.4	31	42.2	7.6	2.3	41.8	8.3	3.5	
Western Corn Belt Plains	9.2.3	26	16.0	2.8	2.2	16.0	2.8	2.2	
Cross Timbers	9.4.5	26	7.9	3.7	1.4	7.9	4.2	3.3	
Eastern Cascades Slopes and Foothills	6.2.8	27	19.7	4.7	0.5	21.5	6.6	3.6	
Arizona/New Mexico Mountains	13.1.1	25	18.4	9.1	7.8	20.3	11.7	10.1	
Willamette Valley	7.1.9	24	59.2	23.5	5.5	65.2	25.5	8.3	
Boston Mountains	8.4.6	23	15.9	7.1	3.4	15.9	7.1	3.4	
Southern and Baja California Pine-Oak Mountains	11.1.3	22	25.8	4.5	0.5	25.2	3.4	1.1	
Central Irregular Plains	9.2.4	21	11.3	2.2	1.5	11.3	2.2	1.5	
California Coastal Sage, Chaparral, and Oak Woodlands	11.1.1	21	33.4	2.4	1.4	34.9	4.6	2.7	
Northern Basin and Range	10.1.3	20	18.1	14.1	5.5	19.1	10.1	3.2	
Mississippi Alluvial Plain	8.5.2	19	8.3	1.1	0.4	8.3	1.1	0.4	
Interior River Valleys and Hills	8.3.2	18	35.3	2.4	1.7	35.3	2.4	1.7	
Driftless Area	8.1.5	15	53.5	22.5	15.4	53.5	22.5	15.4	
Western Gulf Coastal Plain	9.5.1	16	50.3	17.9	7.6	50.4	17.9	7.6	
Central Basin and Range	10.1.5	16	42.4	17.9	1.9	45.4	21.5	8.7	
Eastern Corn Belt Plains	8.2.4	14	10.9	1.7	1.7	10.9	1.7	1.7	
Erie Drift Plain	8.1.10	14	15.4	2.7	0.3	15.4	2.7	0.3	
Mississippi Valley Loess Plains	8.3.6	41	11.6	1.2	0.3	11.6	1.2	0.3	
East Central Texas Plains	8.3.8	10	20.2	5.6	5.6	16.6	1.2	0.3	
Southeastern Wisconsin Till Plains	8.2.1	10	134.9	11.0	10.7	134.9	11.0	10.7	

1 For ecoregions with CLs, minimum/maximum/average total S deposition were 2 summarized. Minimum to maximum range for ecoregion total S deposition was 0.32 – 32.20 Kg 3 S/ha-yr for 2001-2003 and 0.27 - 7.59 Kg S/ha-yr for 2018 - 2020. Average values ranged from 4 1.77 to 8.63 S/ha-yr for 2018-2020 to 2001-2003, respectively (Table 5A-12). Table 5A-13 5 shows the number of ecoregions with <2, 2-5, 5-7, 7-10, >10 Kg S/ha-yr. For the period 2001-6 2003, 16 ecoregions had an average total S deposition over 10 Kg S/ha-yr while there were none 7 in the period 2018-2020. Median S deposition in Kg S/ha-yr are summarized in Table 5A-14 8 and 5A-15. in Ecoregions with the highest average total S deposition were Western Allegheny 9 Plateau, Erie Drift Plain, North Central Appalachians, Central Appalachians, Northern Piedmont, 10 Eastern Corn Belt Plains, Southwestern Appalachians, and Ridge and Valley, all in the Mid-11 Atlantic region of the eastern U.S (Table 5A-14 and 5A-15). 12

#### 1 Table 5A-12. Summary of total S deposition (Kg S/ha-yr) estimates (based on TDEP) at CL locations for 69 ecoregions with at least one CL.

	Total Sulfur deposition (Kg S/ha-yr)										
	2001-03	2006-08	2010-12	2014-16	2018-20						
Minimum	0.32	0.31	0.36	0.52	0.27						
Maximum	32.20	25.97	12.75	9.38	7.59						
Average	8.63	7.39	3.76	2.55	1.77						

Summary of the number of ecoregions with median deposition in the range 3 Table 5A-13. 4 of <2, 2-5, 5-7, 7-10, >10 Kg S/ha-yr for the 84 ecoregions determined by 5 GIS zonal statistic. Deposition based on TDEP.

Total Sulfur Deposition	Number of ecoregions per deposition class									
kg S/ha-yr	2001-03	2006-08	2010-12	2014-16	2018-20					
>10	16	11	0	0	0					
7-10	10	10	5	0	0					
5-7	11	14	10	0	0					
2-5	13	14	31	45	33					
<2	34	35	38	39	51					

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Table 5A-14. Median total sulfur deposition (Kg S/ha-yr) of deposition estimates (based on TDEP) across CL locations for 69 ecoregions with at least one CL. **Deposition based on TDEP.** 

Ecorogian Nama	Codo		No.	2001-03	2006-08	2010-12	2014-16	2018-20
Ecoregion Name	Code		CLs	(kg/ha-yr)	(kg/ha-yr)	(kg/ha-yr)	(kg/ha-yr)	(kg/ha-yr)
Columbia Plateau	10.1.2	W	2	0.8436	0.8335	0.7607	0.6267	0.3812
Northern Basin and Range	10.1.3	W	20	0.9258	1.0470	1.0229	1.0412	0.7504
Wyoming Basin	10.1.4	W	3	0.7697	0.7580	0.6998	0.6846	0.5928
Central Basin and Range	10.1.5	W	16	0.8596	0.6641	0.6670	0.7573	0.5654
Colorado Plateaus	10.1.6	W	1	1.3223	1.4380	1.2534	1.3313	0.8433
Snake River Plain	10.1.8	W	2	0.7953	0.9315	0.9799	0.7949	0.5494
Southern and Central California	11.1.1	W	21	1.6483	1.2030	1.2634	0.9755	1.0646
Central California Valley	11.1.2	W	2	2.1744	1.7039	1.5387	1.4604	1.1912
Southern California Mountains	11.1.3	W	22	1.4499	1.2130	1.2395	1.0407	0.8588
Arizona/New Mexico Mountains	13.1.1	W	25	2.0662	2.5767	1.9619	1.4718	0.8100
Northern Lakes and Forests	5.2.1	Е	839	4.0127	3.1001	2.3408	1.8413	1.3135
Northern Minnesota Wetlands	5.2.2	Е	2	2.1930	2.2114	1.5100	1.1989	0.9099
Northeastern Highlands	5.3.1	Е	2851	7.2925	6.1223	3.1183	2.2176	1.4778
North Central Appalachians	5.3.3	Е	216	15.7250	13.3726	5.8302	3.1733	2.1716
Middle Rockies	6.2.10	W	496	1.4822	1.5250	1.3332	1.0605	0.8719
Klamath Mountains	6.2.11	W	81	0.9200	1.0682	1.0603	0.9866	0.8436
Sierra Nevada	6.2.12	W	353	1.4035	1.2449	1.2725	1.1730	1.0142
Wasatch and Uinta Mountains	6.2.13	W	96	1.7506	1.9154	1.6417	1.7174	1.1053

Ecorogion Name			No.	2001-03	2006-08	2010-12	2014-16	2018-20
	Code	E/VV	CLs	(kg/ha-yr)	(kg/ha-yr)	(kg/ha-yr)	(kg/ha-yr)	(kg/ha-yr)
Southern Rockies	6.2.14	W	372	1.6341	1.7004	1.2931	1.1036	0.7382
Idaho Batholith	6.2.15	W	188	1.2067	1.5204	1.3871	1.1361	0.7188
Northern Rockies	6.2.3	W	86	1.1757	1.2198	1.0150	0.9288	0.6187
Canadian Rockies	6.2.4	W	31	1.2702	1.4281	1.0784	0.9907	0.7898
North Cascades	6.2.5	W	162	1.9444	1.8263	1.4712	1.4767	1.1931
Cascades	6.2.7	W	179	1.2474	1.5117	1.2483	1.2295	1.0687
Eastern Cascades Slopes and Foothills	6.2.8	W	27	0.6646	0.7499	0.7278	0.7420	0.6175
Blue Mountains	6.2.9	W	63	0.6261	0.6766	0.7181	0.8533	0.4616
Puget Lowland	7.1.7	W	38	2.2794	1.9380	1.5475	2.2492	1.3561
Coast Range	7.1.8	W	115	2.4906	2.3092	2.0744	2.0867	1.5174
Willamette Valley	7.1.9	W	24	1.7052	1.4381	1.4547	1.7550	1.0785
Eastern Great Lakes Lowlands	8.1.1	E	83	8.0352	6.5002	3.2588	2.1575	1.4427
Erie Drift Plain	8.1.10	E	14	18.6152	15.4885	7.8262	5.1417	2.8388
Northern Allegheny Plateau	8.1.3	E	199	11.6864	10.4483	4.6870	2.7008	1.7257
North Central Hardwood Forests	8.1.4	E	94	5.2991	3.7229	2.8589	2.1240	1.4780
Driftless Area	8.1.5	E	15	6.1584	5.3369	3.5647	2.7563	2.1083
Southern Michigan/Northern Indiana	8.1.6	Е	33	10.3614	8.9903	5.4132	3.3507	2.3733
Northeastern Coastal Zone	8.1.7	Е	565	9.2880	8.2813	3.7122	2.3007	1.9083
Acadian Plains and Hills	8.1.8	Е	494	4.9838	5.4237	2.8289	1.9501	1.4418
Southeastern Wisconsin Till Plains	8.2.1	Е	10	6.9406	5.7085	3.9255	2.7417	1.9581
Central Corn Belt Plains	8.2.3	E	2	10.6435	9.7895	5.9799	4.4423	2.5047
Eastern Corn Belt Plains	8.2.4	Е	14	17.4280	13.4790	7.9019	4.7601	2.8735
Northern Piedmont	8.3.1	Е	231	15.1825	12.9379	5.6325	3.3300	2.2078
Interior River Valleys and Hills	8.3.2	Е	18	12.5930	11.0339	6.5440	4.2474	2.9442
Interior Plateau	8.3.3	Е	71	13.1051	9.8406	5.5813	4.0054	2.7395
Piedmont	8.3.4	Е	508	12.2634	10.1418	4.2405	2.6855	2.0337
Southeastern Plains	8.3.5	Е	390	10.8772	9.1412	4.8250	3.4937	2.4093
Mississippi Valley Loess Plains	8.3.6	E	41	9.4020	7.6610	4.7177	4.4386	3.5668
South Central Plains	8.3.7	Е	153	7.7690	7.1450	5.0275	4.6912	3.8806
East Central Texas Plains	8.3.8	E	10	6.3592	6.3713	4.6506	4.7800	3.7907
Ridge and Valley	8.4.1	E	1292	14.1834	11.9342	5.7140	3.3291	1.9421
Central Appalachians	8.4.2	Е	372	17.0275	13.9833	7.2537	4.0873	2.4291
Western Allegheny Plateau	8.4.3	Е	35	17.0756	14.1224	7.5947	4.1935	2.5646
Blue Ridge	8.4.4	E	1972	11.2890	9.5751	4.4112	2.6974	2.0560
Ozark Highlands	8.4.5	Е	56	6.9470	6.1841	4.8676	3.2446	2.6634
Boston Mountains	8.4.6	E	23	6.2465	5.8956	4.6014	3.4302	2.7833
Arkansas Valley	8.4.7	Е	31	5.7040	5.3761	4.2414	3.3530	2.9080
Ouachita Mountains	8.4.8	Е	42	6.0931	5.7076	4.6545	4.0534	3.5802
Southwestern Appalachians	8.4.9	Е	117	17.2703	14.4428	5.5887	4.1742	2.9277
Middle Atlantic Coastal Plain	8.5.1	Е	105	14.1038	12.0745	5.3535	3.5822	2.4066
Mississippi Alluvial Plain	8.5.2	Е	19	7.0222	5.4477	4.0627	3.6724	3.0482
Southern Coastal Plain	8.5.3	Е	142	8.7020	5.9155	4.5581	4.1848	3.3530

Ecorogian Nama	Codo		No.	2001-03	2006-08	2010-12	2014-16	2018-20
	Code		CLs	(kg/ha-yr)	(kg/ha-yr)	(kg/ha-yr)	(kg/ha-yr)	(kg/ha-yr)
Atlantic Coastal Pine Barrens	8.5.4	E	234	13.8762	12.0097	5.4040	3.8876	2.8384
Western Corn Belt Plains	9.2.3	E	26	4.7163	4.0139	2.8494	2.3457	1.9949
Central Irregular Plains	9.2.4	Е	21	5.5499	5.1167	3.9854	2.9482	2.2863
Northwestern Glaciated Plains	9.3.1	Е	2	0.6676	0.7441	0.5393	0.5628	0.4593
Central Great Plains	9.4.2	E	5	4.3246	4.6747	2.8565	2.7344	2.4439
Flint Hills	9.4.4	Е	7	4.4474	4.3621	2.9061	2.5660	2.2734
Cross Timbers	9.4.5	Е	26	4.8935	4.4674	3.2464	3.1676	2.7247
Texas Blackland Prairies	9.4.7	E	3	6.5071	5.9520	4.4689	4.3720	3.6600
Western Gulf Coastal Plain	9.5.1	Е	16	7.5945	6.9910	4.9189	5.3084	4.3375

# Table 5A-15.Median sulfur deposition (Kg S/ha-yr) for the 84 ecoregions determined by<br/>GIS zonal statistic. Deposition based on TDEP.

Ecoregion III		Total Median Sulfur Deposition (Kg S/ha-yr)							
Name	Code	2001-03	2006-08	2010-12	2014-16	2018-20			
Columbia Plateau	10.1.2	0.4575	0.423	0.4339	0.5046	0.2873			
Northern Basin and Range	10.1.3	0.3447	0.3736	0.5295	0.4769	0.2872			
Wyoming Basin	10.1.4	0.6354	0.673	0.5232	0.5582	0.4235			
Central Basin and Range	10.1.5	0.4931	0.4465	0.4682	0.5229	0.3402			
Colorado Plateaus	10.1.6	0.7419	0.7448	0.5604	0.6091	0.3206			
Arizona/New Mexico Plateau	10.1.7	0.8232	0.801	0.6367	0.5653	0.3321			
Snake River Plain	10.1.8	0.4799	0.6588	0.5869	0.5938	0.381			
Mojave Basin and Range	10.2.1	0.5775	0.4134	0.4165	0.4087	0.2954			
Chihuahuan Deserts	10.2.10	1.2098	1.1233	1.1098	1.2169	0.8642			
Sonoran Basin and Range	10.2.2	0.5358	0.4614	0.45	0.4381	0.2889			
Southern and Central California Chaparral and Oak Woodlands	11.1.1	1.12	0.9523	0.9446	0.8409	0.7363			
Central California Valley	11.1.2	1.0886	0.9161	0.8184	0.7977	0.6555			
Southern California Mountains	11.1.3	1.2321	1.0846	1.0668	0.9797	0.8343			
Madrean Archipelago	12.1.1	1.1561	1.144	0.9214	0.9407	0.4895			
Arizona/New Mexico Mountains	13.1.1	1.4323	1.4098	1.1677	1.0302	0.5954			
Southern Florida Coastal Plain	15.4.1	5.9639	5.1638	4.199	4.3358	3.7554			
Northern Lakes and Forests	5.2.1	4.2923	3.2398	2.4385	1.8937	1.3265			
Northern Minnesota Wetlands	5.2.2	2.2819	2.1153	1.4503	1.1332	0.8588			
Northeastern Highlands	5.3.1	6.457	5.7779	3.0112	1.9925	1.3362			
North Central Appalachians	5.3.3	18.0836	15.0511	7.2364	4.0916	2.4012			
Middle Rockies	6.2.10	1.0399	1.1392	0.9349	0.8577	0.7085			
Klamath Mountains	6.2.11	0.9013	1.0525	1.022	1.0653	0.925			
Sierra Nevada	6.2.12	1.3177	1.1446	1.2417	1.141	0.9773			
Wasatch and Uinta Mountains	6.2.13	1.3604	1.3833	1.1818	1.2713	0.7741			
Southern Rockies	6.2.14	1.1427	1.1821	0.9241	0.8462	0.5419			
Idaho Batholith	6.2.15	0.8952	1.1615	1.0974	0.9326	0.5975			

Ecoregion III	Total Median Sulfur Deposition (Kg S/					a-yr)
Name	Code	2001-03	2006-08	2010-12	2014-16	2018-20
Northern Rockies	6.2.3	0.8969	0.9757	0.829	0.7912	0.5242
Canadian Rockies	6.2.4	1.2218	1.348	0.9682	0.97	0.7759
North Cascades	6.2.5	1.643	1.5503	1.28	1.3852	1.0878
Cascades	6.2.7	1.6874	1.662	1.4078	1.506	1.2381
Eastern Cascades Slopes and Foothills	6.2.8	0.4394	0.4902	0.4704	0.5498	0.471
Blue Mountains	6.2.9	0.4603	0.5022	0.523	0.6145	0.3557
Puget Lowland	7.1.7	2.1291	1.6318	1.3687	2.1064	1.2462
Coast Range	7.1.8	2.386	2.1428	1.9998	2.0311	1.5004
Willamette Valley	7.1.9	1.6079	1.4789	1.4314	1.7067	1.0764
Eastern Great Lakes Lowlands	8.1.1	10.9736	8.8203	4.0355	2.7074	1.6382
Erie Drift Plain	8.1.10	18.3931	15.104	8.0732	4.9947	2.8124
Northern Allegheny Plateau	8.1.3	11.9249	10.2437	4.8068	2.7891	1.6814
North Central Hardwood Forests	8.1.4	4.5736	3.4207	2.6262	2.0107	1.3864
Driftless Area	8.1.5	5.3867	4.9968	3.3673	2.6069	1.9468
Southern Michigan/Northern Indiana Drift Plains	8.1.6	9.6189	8.3446	5.3163	3.2534	2.161
Northeastern Coastal Zone	8.1.7	9.5712	8.4241	3.8242	2.3994	1.8734
Acadian Plains and Hills	8.1.8	4.459	4.6136	2.3792	1.6499	1.2222
Southeastern Wisconsin Till Plains	8.2.1	7.0223	6.3692	3.977	2.7375	2.0215
Huron/Erie Lake Plains	8.2.2	9.8648	8.5901	5.2196	3.1513	2.1087
Central Corn Belt Plains	8.2.3	9.7812	8.9628	5.4209	4.1072	2.4466
Eastern Corn Belt Plains	8.2.4	14.8424	11.9827	7.0763	4.1088	2.5906
Northern Piedmont	8.3.1	14.9402	12.5778	5.3007	3.3249	2.1241
Interior River Valleys and Hills	8.3.2	10.5462	9.3038	6.2016	4.2898	3.03
Interior Plateau	8.3.3	13.5173	10.958	6.2411	4.1623	2.7293
Piedmont	8.3.4	11.7106	9.5824	4.3372	2.6194	1.8919
Southeastern Plains	8.3.5	9.6797	8.051	4.3423	3.4774	2.6347
Mississippi Valley Loess Plains	8.3.6	8.6391	6.6922	4.5953	3.9633	3.176
South Central Plains	8.3.7	7.3432	6.7813	4.9139	4.7025	3.6386
East Central Texas Plains	8.3.8	6.4132	5.1437	3.8214	4.4478	3.6219
Ridge and Valley	8.4.1	14.0986	11.8557	5.3063	3.2326	2.1381
Central Appalachians	8.4.2	16.2032	13.2761	7.0458	4.1165	2.3152
Western Allegheny Plateau	8.4.3	20.3503	16.3579	8.2608	4.7637	2.887
Blue Ridge	8.4.4	11.1204	9.2557	4.4061	2.6059	1.9493
Ozark Highlands	8.4.5	6.3057	5.8383	4.6492	3.1904	2.5927
Boston Mountains	8.4.6	5.9764	5.7193	4.4839	3.3348	2.7899
Arkansas Valley	8.4.7	5.5359	5.2017	4.1517	3.3783	2.9723
Ouachita Mountains	8.4.8	6.1981	5.8193	4.6709	4.0934	3.5246
Southwestern Appalachians	8.4.9	14.7077	11.5575	5.4669	3.4563	2.6107
Middle Atlantic Coastal Plain	8.5.1	10.5248	9.3374	5.0943	3.426	2.3606
Mississippi Alluvial Plain	8.5.2	7.3738	6.0578	4.2173	3.9128	3.1692

Ecoregion III		Total Median Sulfur Deposition (Kg S/ha-yr)									
Name	Code	2001-03	2006-08	2010-12	2014-16	2018-20					
Southern Coastal Plain	8.5.3	7.9369	6.0166	4.4339	3.9496	3.232					
Atlantic Coastal Pine Barrens	8.5.4	14.0323	12.2726	5.6121	3.8026	2.747					
Northern Glaciated Plains	9.2.1	2.0376	2.084	1.7364	1.3281	1.2238					
Lake Agassiz Plain	9.2.2	1.9749	1.9895	1.4408	1.1922	1.066					
Western Corn Belt Plains	9.2.3	4.521	4.2523	2.9805	2.5558	1.9268					
Central Irregular Plains	9.2.4	5.8102	5.3365	4.1299	2.9768	2.2734					
Northwestern Glaciated Plains	9.3.1	1.5672	1.6233	1.3799	1.2046	1.0888					
Northwestern Great Plains	9.3.3	1.2028	1.3341	1.0109	0.8764	0.8163					
Nebraska Sand Hills	9.3.4	1.6666	1.991	1.4768	1.3589	1.3629					
High Plains	9.4.1	1.6	1.5156	1.2682	1.3302	0.983					
Central Great Plains	9.4.2	3.058	2.9854	2.1563	2.1892	1.8397					
Southwestern Tablelands	9.4.3	1.3027	1.2442	0.9851	1.116	0.6471					
Flint Hills	9.4.4	4.4375	4.0337	2.8507	2.4599	1.9342					
Cross Timbers	9.4.5	4.5832	3.9558	3.0153	3.0523	2.6092					
Edwards Plateau	9.4.6	3.0692	2.7607	2.211	2.5378	2.1013					
Texas Blackland Prairies	9.4.7	6.1482	4.8692	3.8494	4.0241	3.3937					
Western Gulf Coastal Plain	9.5.1	6.9462	5.6366	4.3126	4.7353	4.3323					
Southern Texas Plains	9.6.1	3.722	3.0284	2.5427	3.0882	2.3617					

### 5A.2.2.1 Ecoregion Critical Load Exceedances – Sulfur Only

2 Critical load exceedances were evaluated for the 69 ecoregions that had at least one CL. 3 Of the 69, 58 ecoregions had 10 or more values, and were therefore used in this analysis. We 4 evaluated both All CLs and only positive values in the Ecoregion. Exceedances were evaluated 5 with respect to 2001-2003, 2006-2008, 2012-2014, 2014-2016, and 2018-2020 for S only and 6 combined N and/or S deposition. Exceedances were calculated for ANC thresholds of 20, 30, 50 7  $\mu$ eq/L and combined 50  $\mu$ eq/L in the East and 20  $\mu$ eq/L in the West (Noted as 50/20  $\mu$ eq/L). 8 Results of S only exceed are summarized in Table 5A-16 for each endpoint and time period. 9 Results of S only exceedances are included in Tables 5A-17 and 5A-24. Results for N and S 10 deposition were not summarized into tables. See section titled "Critical Load Exceedances" for a 11 description of how exceedances were calculated.

12 This section describes the results for ecoregion CL exceedances considering S deposition 13 only (Tables 5A-7 to 5A-24 and Figures 5A-32 to 5A-43). A summary of ecoregion CLs and

exceedance results can be found in Table 5A-16 for each of the ANC thresholds and deposition

15 time periods. In addition, ecoregion averages, 5th and 95th percentiles and counts of EXs >5,

16 10, 15, and 25 are summarized for positive CLs.

Exceedances were low for the 58 ecoregions using ANC thresholds of 20 and 30 μeq/L
for the most recent years (2018-2020 and 2014-2016). For the 58 ecoregions, 40 and 48 had no

- 1 exceedances at all for 2018-2020 and 2014-2016, respectively. Of the remaining 29 and 21
- 2 ecoregions, only 6 and 9 had greater than 5% exceedance and 3 and 5 had greater than 10% for
- 3 ANC thresholds of 20  $\mu$ eq/L for the two deposition periods. For the following three deposition
- 4 periods 2010-2012, 2006-2008, and 2001-2003, the number of ecoregions without an exceedance
- 5 decreased to 35, 31, and 29 while the number with greater than 10% exceedance increased to 8,
- 6 21, and 23, respectively Critical loads based on an ANC threshold of 30 μeq/L had slightly
- 7 higher exceedances across all deposition periods. The Southeastern Plans, Southern Coastal
- 8 Plain, and Atlantic Coastal Pine Barrens are ecoregions known to have naturally acidic surface
- 9 waters and the high exceedances calculated for these ecoregions are likely not driven by air
- 10 pollution deposition but instead by natural acidity linked to DOC, hydrology, and natural
- 11 biogeochemical processes (Stauffer and Canfield Jr. 1992). Central Appalachians Acadian
- 12 Plains and Hills Northern Appalachian and Atlantic Maritime Highlands are ecoregions that have
- 13 documented deposition driven acidification.

Table 5A-16.Summary of Ecoregion results for critical load (CL) exceedances (EX) for<br/>each ANC threshold and time periods for the 58 Ecoregions with 10 or<br/>more values. The range of CL and Sulfur (S) deposition in Kg S/ha/yr<br/>represents values that exceed the CL. Average percent EX represents the<br/>average EX found in the 58 ecoregions for a particular ANC threshold and<br/>time period. The four numbers (#) represent the number of ecoregions that<br/>have greater than 5%, 10%, 15%, and 25% EX within the ecoregion for a<br/>given ANC threshold and time period.

	Critical Load	S Deposition					
Time Period	Average (5th to 95th)	Average (5th to 95th)	Ave. % EX	# >5% EX	# >10% EX	# >15% EX	#>25% EA
		ANC Threshol	d = 50/20	µeq/L			
2018-2020	1 (0.1-2.4)	2.1 (1.2-3.6)	3.1	12	7	2	0
2014-2016	1.5 (0.2-3.4)	3 (1.7-5)	4.0	16	8	3	0
2010-2012	2.4 (0.2-5.4)	2.4 (0.2-5.4)	6.1	24	16	7	3
2006-2008	4.9 (0.5-11.2)	9.5 (3.3-15)	12.1	33	24	20	10
2001-2003	5.7 (0.5-13)	11.4 (4.3-17.9)	14.4	33	27	20	13
		ANC Thresh	old = 50 µ	eq/L			
2018-2020	1 (0.1-2.4)	2.1 (1-3.6)	3.2	12	7	0	0
2014-2016	1.5 (0.2-3.4)	3 (1.5-5)	4.3	16	8	3	0
2010-2012	2.3 (0.2-5.4)	4.5 (2-7.7)	6.3	25	16	7	3
2006-2008	4.9 (0.4-11.1)	9.4 (3.1-15)	12.4	33	24	20	10
2001-2003	5.6 (0.5-12.9)	11.3 (4-17.9)	14.6	33	27	20	13
		ANC Thresh	old = 30 µ	eq/L			
2018-2020	3.2 (1.4-6.2)	2 (0.9-3.4)	1.9	5	4	1	0
2014-2016	1.7 (0.2-3.9)	3.1 (1.3-5.3)	3.0	11	5	2	1
2010-2012	2.7 (0.3-5.7)	4.6 (1.7-7.8)	4.7	20	11	4	1
2006-2008	5.6 (0.8-11.5)	9.7 (3.2-15.1)	10.3	30	23	17	8
2001-2003	6.6 (0.9-13.7)	11.5 (4.2-17.9)	13.1	31	24	20	12
		Critical Load for	· ANC = 20	) µeq/L			
2018-2020	3.8 (1.8-7.1)	2 (0.9-3.4)	1.5	6	3	1	0
2014-2016	1.9 (0.3-4.1)	3.4 (1.5-5.4)	2.7	8	4	3	2
2010-2012	2.9 (0.5-6.1)	4.8 (2.1-8.1)	4.2	17	7	3	2
2006-2008	6 (1.1-11.8)	9.8 (4.1-15.2)	9.4	27	20	17	8
2001-2003	7.1 (1.3-14.1)	11.8 (4.5-18.1)	12.3	28	22	21	11

9 Critical loads determined for ANC thresholds of 50 and 50/20 µeq/L had higher percent
10 exceedances within ecoregions and more ecoregions with some level of exceedances particularly
11 for the early deposition periods of 2010-2012, 2006-2008, and 2001-2003 (Tables 5A-19 to 5A12 23 and Figures 5A-39 to 5A-44). For critical loads using an ANC threshold of 50 µeq/L, 31, 25,
13 21, 21, and 21 of the 58 Ecoregions had no critical load exceedances for the 5 deposition periods
14 2018-2020, 2014-2016, 2010-2012, 2006-2008, and 2001-2003. Of the remaining Ecoregions,

- 1 13,17, 36, 43, and 44 had greater than 5% exceedances and 8, 9, 25, 33, and 35 Ecoregions had
- 2 exceedance percentage greater than 10%.

	Sulfur only - ANC = 20 ueq/L										
	No. = 69					% Exc	eedance	s			
Ecoregion			CI	_≤0	201	18-20	20	14-16			
Name	#	No.	No.	%	All	CL>0	All	CL>0			
Northern Appalachian and Atlantic Maritime Highlands	5.3.1	2851	11	0.4	1.6	1.2	2.7	2.4			
Blue Ridge	8.4.4	1972	3	0.2	0.9	0.7	1.8	1.6			
Ridge and Valley	8.4.1	1292	2	0.2	0.9	0.8	4.7	4.6			
Northern Lakes and Forests	5.2.1	839	1	0.1	0.5	0.4	1.2	1.1			
Northeastern Coastal Zone	8.1.7	565	1	0.2	0.2	0.0	0.9	0.7			
Piedmont	8.3.4	508	0	0.0	0.2	0.2	0.6	0.6			
Middle Rockies	6.2.10	496	0	0.0	0.2	0.2	0.6	0.6			
Acadian Plains and Hills	8.1.8	494	2	0.4	2.0	1.6	2.6	2.2			
Southeastern Plains	8.3.5	390	3	0.8	4.9	4.1	6.9	6.2			
Central Appalachians	8.4.2	372	4	1.1	3.8	2.7	5.6	4.6			
Southern Rockies	6.2.14	372	1	0.3	0.5	0.3	1.9	1.6			
Sierra Nevada	6.2.12	353	11	3.1	4.2	1.1	5.9	2.8			
Atlantic Coastal Pine Barrens	8.5.4	234	1	0.4	12.4	12.0	17.9	17.5			
Northern Piedmont	8.3.1	231	0	0.0	0.0	0.0	0.4	0.4			
North Central Appalachians	5.3.3	216	0	0.0	0.0	0.0	2.3	2.3			
Northern Allegheny Plateau	8.1.3	199	0	0.0	0.5	0.5	1.0	1.0			
Idaho Batholith	6.2.15	188	1	0.5	0.5	0.0	0.5	0.0			
Cascades	6.2.7	179	4	2.2	3.9	1.7	5.0	2.8			
North Cascades	6.2.5	162	1	0.6	0.6	0.0	0.6	0.0			
South Central Plains	8.3.7	153	2	1.3	9.8	8.5	12.4	11.1			
Southern Coastal Plain	8.5.3	142	1	0.7	20.4	19.7	27.5	26.8			
Southwestern Appalachians	8.4.9	117	0	0.0	0.0	0.0	0.0	0.0			
Coast Range	7.1.8	115	0	0.0	0.0	0.0	0.0	0.0			
Middle Atlantic Coastal Plain	8.5.1	105	1	1.0	2.9	1.9	4.8	3.8			
Wasatch and Uinta Mountains	6.2.13	96	0	0.0	0.0	0.0	0.0	0.0			
North Central Hardwood Forests	8.1.4	94	0	0.0	0.0	0.0	0.0	0.0			
Columbia Mountains/Northern Rockies	6.2.3	86	1	1.2	1.2	0.0	2.3	1.2			
Eastern Great Lakes Lowlands	8.1.1	83	1	1.2	1.2	0.0	1.2	0.0			
Klamath Mountains	6.2.11	81	0	0.0	0.0	0.0	0.0	0.0			
Interior Plateau	8.3.3	71	0	0.0	0.0	0.0	0.0	0.0			
Blue Mountains	6.2.9	63	0	0.0	0.0	0.0	0.0	0.0			
Ozark Highlands	8.4.5	56	0	0.0	0.0	0.0	1.8	1.8			

# Table 5A-17. Percent Ecoregion Exceedances of aquatic CLs for Sulfur only by ANC threshold of 20 μeq/L for deposition years of 2018-20 and 2014-16.

	Sulfur only - ANC = 20 ueq/L									
	No. = 69					% Exc	eedance	S		
Ecoregion			CI	_≤0	201	8-20	20	14-16		
Name	#	No.	No.	%	All	CL>0	All	CL>0		
Ouachita Mountains	8.4.8	42	0	0.0	0.0	0.0	0.0	0.0		
Mississippi Valley Loess Plains	8.3.6	41	0	0.0	2.4	2.4	9.8	9.8		
Strait of Georgia/Puget Lowland	7.1.7	38	0	0.0	0.0	0.0	0.0	0.0		
Western Allegheny Plateau	8.4.3	35	0	0.0	0.0	0.0	0.0	0.0		
Southern Michigan/Northern Indiana Drift Plains	8.1.6	33	0	0.0	0.0	0.0	3.0	3.0		
Arkansas Valley	8.4.7	31	0	0.0	0.0	0.0	0.0	0.0		
Canadian Rockies	6.2.4	31	0	0.0	0.0	0.0	0.0	0.0		
Eastern Cascades Slopes and Foothills	6.2.8	27	0	0.0	0.0	0.0	0.0	0.0		
Cross Timbers	9.4.5	26	0	0.0	0.0	0.0	0.0	0.0		
Western Corn Belt Plains	9.2.3	26	0	0.0	0.0	0.0	0.0	0.0		
Arizona/New Mexico Mountains	13.1.1	25	0	0.0	0.0	0.0	0.0	0.0		
Willamette Valley	7.1.9	24	0	0.0	0.0	0.0	0.0	0.0		
Boston Mountains	8.4.6	23	0	0.0	0.0	0.0	0.0	0.0		
Southern and Baja California Pine-Oak Mountains	11.1.3	22	0	0.0	0.0	0.0	4.5	4.5		
Central Irregular Plains	9.2.4	21	0	0.0	0.0	0.0	0.0	0.0		
California Coastal Sage, Chaparral, and Oak Woodlands	11.1.1	21	0	0.0	0.0	0.0	0.0	0.0		
Northern Basin and Range	10.1.3	20	0	0.0	0.0	0.0	0.0	0.0		
Mississippi Alluvial Plain	8.5.2	19	0	0.0	5.3	5.3	5.3	5.3		
Interior River Valleys and Hills	8.3.2	18	0	0.0	0.0	0.0	0.0	0.0		
Western Gulf Coastal Plain	9.5.1	16	0	0.0	0.0	0.0	0.0	0.0		
Central Basin and Range	10.1.5	16	0	0.0	0.0	0.0	0.0	0.0		
Driftless Area	8.1.5	15	0	0.0	0.0	0.0	0.0	0.0		
Erie Drift Plain	8.1.10	14	0	0.0	0.0	0.0	0.0	0.0		
Eastern Corn Belt Plains	8.2.4	14	0	0.0	0.0	0.0	0.0	0.0		
East Central Texas Plains	8.3.8	10	0	0.0	10.0	10.0	10.0	10.0		
Southeastern Wisconsin Till Plains	8.2.1	10	0	0.0	0.0	0.0	0.0	0.0		
Flint Hills	9.4.4	7	0	0.0	14.3	14.3	28.6	28.6		
Central Great Plains	9.4.2	5	0	0.0	0.0	0.0	0.0	0.0		
Wyoming Basin	10.1.4	3	0	0.0	0.0	0.0	0.0	0.0		
Texas Blackland Prairies	9.4.7	3	0	0.0	0.0	0.0	0.0	0.0		
Northern Minnesota Wetlands	5.2.2	2	0	0.0	0.0	0.0	50.0	50.0		
Snake River Plain	10.1.8	2	0	0.0	0.0	0.0	0.0	0.0		
Central California Valley	11.1.2	2	0	0.0	0.0	0.0	0.0	0.0		
Columbia Plateau	10.1.2	2	0	0.0	0.0	0.0	0.0	0.0		
Central Corn Belt Plains	8.2.3	2	0	0.0	0.0	0.0	0.0	0.0		
Northwestern Glaciated Plains	9.3.1	2	0	0.0	0.0	0.0	0.0	0.0		
Colorado Plateaus	10.1.6	1	0	0.0	0.0	0.0	0.0	0.0		

Table 5A-18.Percent Ecoregion Exceedances of aquatic CLs for Sulfur only by ANC<br/>threshold of 20 µeq/L for deposition years of 2010-12, 2006-08 and 2001-03.

	Sulfur only - ANC = 20 ueq/L										
	No. = 69					C	% Exce	edances	;		
Ecoregion	I		C	L≤0	2010-	-2012	2006	-2008	2001	-2003	
Name	#	No.	No.	%	All	CL>0	All	CL>0	All	CL>0	
Northern Appalachian and Atlantic Maritime Highlands	5.3.1	2851	11	0.4	4.5	4.1	14.9	14.5	19.9	19.5	
Blue Ridge	8.4.4	1972	3	0.2	6.1	5.9	32.3	32.2	43.7	43.5	
Ridge and Valley	8.4.1	1292	2	0.2	9.8	9.6	29.3	29.2	38.5	38.3	
Northern Lakes and Forests	5.2.1	839	1	0.1	2.9	2.7	8.8	8.7	16.0	15.9	
Northeastern Coastal Zone	8.1.7	565	1	0.2	2.1	1.9	8.3	8.1	10.1	9.9	
Piedmont	8.3.4	508	0	0.0	1.8	1.8	11.8	11.8	16.1	16.1	
Middle Rockies	6.2.10	496	0	0.0	0.8	0.8	0.8	0.8	0.8	0.8	
Acadian Plains and Hills	8.1.8	494	2	0.4	4.7	4.3	10.5	10.1	10.1	9.7	
Southeastern Plains	8.3.5	390	3	0.8	9.5	8.7	20.5	19.7	24.1	23.3	
Central Appalachians	8.4.2	372	4	1.1	15.9	14.8	44.4	43.3	53.8	52.7	
Southern Rockies	6.2.14	372	1	0.3	2.2	1.9	2.7	2.4	2.7	2.4	
Sierra Nevada	6.2.12	353	11	3.1	5.1	2.0	5.1	2.0	6.2	3.1	
Atlantic Coastal Pine Barrens	8.5.4	234	1	0.4	23.9	23.5	45.3	44.9	53.8	53.4	
Northern Piedmont	8.3.1	231	0	0.0	1.7	1.7	6.5	6.5	7.8	7.8	
North Central Appalachians	5.3.3	216	0	0.0	5.6	5.6	24.1	24.1	31.5	31.5	
Northern Allegheny Plateau	8.1.3	199	0	0.0	2.0	2.0	7.5	7.5	9.5	9.5	
Idaho Batholith	6.2.15	188	1	0.5	0.5	0.0	0.5	0.0	0.5	0.0	
Cascades	6.2.7	179	4	2.2	3.9	1.7	3.9	1.7	3.9	1.7	
North Cascades	6.2.5	162	1	0.6	0.6	0.0	0.6	0.0	0.6	0.0	
South Central Plains	8.3.7	153	2	1.3	13.7	12.4	19.0	17.6	20.3	19.0	
Southern Coastal Plain	8.5.3	142	1	0.7	28.9	28.2	35.2	34.5	49.3	48.6	
Southwestern Appalachians	8.4.9	117	0	0.0	0.0	0.0	25.6	25.6	41.0	41.0	
Coast Range	7.1.8	115	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	
Middle Atlantic Coastal Plain	8.5.1	105	1	1.0	5.7	4.8	20.0	19.0	24.8	23.8	
Wasatch and Uinta Mountains	6.2.13	96	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	
North Central Hardwood Forests	8.1.4	94	0	0.0	0.0	0.0	0.0	0.0	3.2	3.2	
Columbia Mountains/Northern Rockies	6.2.3	86	1	1.2	3.5	2.3	3.5	2.3	3.5	2.3	
Eastern Great Lakes Lowlands	8.1.1	83	1	1.2	1.2	0.0	2.4	1.2	6.0	4.8	
Klamath Mountains	6.2.11	81	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	
Interior Plateau	8.3.3	71	0	0.0	5.6	5.6	8.5	8.5	12.7	12.7	
Blue Mountains	6.2.9	63	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	
Ozark Highlands	8.4.5	56	0	0.0	3.6	3.6	3.6	3.6	3.6	3.6	
Ouachita Mountains	8.4.8	42	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	
Mississippi Valley Loess Plains	8.3.6	41	0	0.0	12.2	12.2	17.1	17.1	19.5	19.5	
Strait of Georgia/Puget Lowland	7.1.7	38	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	

	Sulfur only - ANC = 20 ueq/L									
	No. = 69					C	% Exce	edances	;	
Ecoregion	•		C	L≤0	2010-	·2012	2006	-2008	2001	-2003
Name	#	No.	No.	%	All	CL>0	All	CL>0	All	CL>0
Western Allegheny Plateau	8.4.3	35	0	0.0	2.9	2.9	20.0	20.0	28.6	28.6
Southern Michigan/Northern Indiana Drift Plains	8.1.6	33	0	0.0	6.1	6.1	15.2	15.2	21.2	21.2
Arkansas Valley	8.4.7	31	0	0.0	3.2	3.2	3.2	3.2	3.2	3.2
Canadian Rockies	6.2.4	31	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Eastern Cascades Slopes and Foothills	6.2.8	27	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Cross Timbers	9.4.5	26	0	0.0	0.0	0.0	3.8	3.8	7.7	7.7
Western Corn Belt Plains	9.2.3	26	0	0.0	0.0	0.0	0.0	0.0	3.8	3.8
Arizona/New Mexico Mountains	13.1.1	25	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Willamette Valley	7.1.9	24	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Boston Mountains	8.4.6	23	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Southern and Baja California Pine-Oak Mountains	11.1.3	22	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Central Irregular Plains	9.2.4	21	0	0.0	0.0	0.0	4.8	4.8	4.8	4.8
California Coastal Sage, Chaparral, and Oak Woodlands	11.1.1	21	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Northern Basin and Range	10.1.3	20	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Mississippi Alluvial Plain	8.5.2	19	0	0.0	5.3	5.3	5.3	5.3	21.1	21.1
Interior River Valleys and Hills	8.3.2	18	0	0.0	5.6	5.6	16.7	16.7	22.2	22.2
Western Gulf Coastal Plain	9.5.1	16	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Central Basin and Range	10.1.5	16	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Driftless Area	8.1.5	15	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Erie Drift Plain	8.1.10	14	0	0.0	7.1	7.1	21.4	21.4	28.6	28.6
Eastern Corn Belt Plains	8.2.4	14	0	0.0	14.3	14.3	28.6	28.6	28.6	28.6
East Central Texas Plains	8.3.8	10	0	0.0	10.0	10.0	10.0	10.0	10.0	10.0
Southeastern Wisconsin Till Plains	8.2.1	10	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Flint Hills	9.4.4	7	0	0.0	28.6	28.6	28.6	28.6	28.6	28.6
Central Great Plains	9.4.2	5	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Wyoming Basin	10.1.4	3	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Texas Blackland Prairies	9.4.7	3	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Northern Minnesota Wetlands	5.2.2	2	0	0.0	50.0	50.0	50.0	50.0	50.0	50.0
Snake River Plain	10.1.8	2	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Central California Valley	11.1.2	2	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Columbia Plateau	10.1.2	2	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Central Corn Belt Plains	8.2.3	2	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Northwestern Glaciated Plains	9.3.1	2	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Colorado Plateaus	10.1.6	1	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0

#### Percent Ecoregion Exceedances of aquatic CLs for Sulfur only by ANC threshold of 30 µeq/L for deposition years of 2018-20 and 2014-16. 1 Table 5A-19.

	Sulfur only - ANC = 30 ueg/L									
	No. = 69					% Exce	edances	3		
Ecoregion			C	L≤0	2018	-2020	2014	-2016		
Name	#	No.	No.	%	All	CL>0	All	CL>0		
Northern Appalachian and Atlantic Maritime Highlands	5.3.1	2851	40	1.4	3.0	1.6	4.9	3.5		
Blue Ridge	8.4.4	1972	13	0.7	2.3	1.6	3.9	3.2		
Ridge and Valley	8.4.1	1292	8	0.6	2.4	1.8	5.8	5.2		
Northern Lakes and Forests	5.2.1	839	1	0.1	1.0	0.8	2.4	2.3		
Northeastern Coastal Zone	8.1.7	565	1	0.2	1.2	1.1	1.6	1.4		
Piedmont	8.3.4	508	1	0.2	1.0	0.8	1.2	1.0		
Middle Rockies	6.2.10	496	4	0.8	1.4	0.6	1.6	0.8		
Acadian Plains and Hills	8.1.8	494	9	1.8	3.8	2.0	4.7	2.8		
Southeastern Plains	8.3.5	390	9	2.3	7.2	4.9	9.5	7.2		
Central Appalachians	8.4.2	372	10	2.7	5.6	3.0	8.3	5.6		
Southern Rockies	6.2.14	372	8	2.2	3.0	0.8	4.0	1.9		
Sierra Nevada	6.2.12	353	29	8.2	11.9	3.7	16.4	8.2		
Atlantic Coastal Pine Barrens	8.5.4	234	9	3.8	17.9	14.1	22.2	18.4		
Northern Piedmont	8.3.1	231	0	0.0	1.3	1.3	1.3	1.3		
North Central Appalachians	5.3.3	216	0	0.0	1.4	1.4	2.3	2.3		
Northern Allegheny Plateau	8.1.3	199	1	0.5	1.0	0.5	1.5	1.0		
Idaho Batholith	6.2.15	188	3	1.6	2.1	0.5	2.7	1.1		
Cascades	6.2.7	179	11	6.1	6.7	0.6	7.3	1.1		
North Cascades	6.2.5	162	1	0.6	0.6	0.0	0.6	0.0		
South Central Plains	8.3.7	153	3	2.0	14.4	12.4	16.3	14.4		
Southern Coastal Plain	8.5.3	142	4	2.8	22.5	19.7	30.3	27.5		
Southwestern Appalachians	8.4.9	117	0	0.0	0.0	0.0	0.0	0.0		
Coast Range	7.1.8	115	0	0.0	0.0	0.0	0.0	0.0		
Middle Atlantic Coastal Plain	8.5.1	105	1	1.0	2.9	1.9	4.8	3.8		
Wasatch and Uinta Mountains	6.2.13	96	0	0.0	0.0	0.0	0.0	0.0		
North Central Hardwood Forests	8.1.4	94	0	0.0	1.1	1.1	1.1	1.1		
Columbia Mountains/Northern Rockies	6.2.3	86	3	3.5	3.5	0.0	3.5	0.0		
Eastern Great Lakes Lowlands	8.1.1	83	1	1.2	1.2	0.0	1.2	0.0		
Klamath Mountains	6.2.11	81	0	0.0	0.0	0.0	0.0	0.0		
Interior Plateau	8.3.3	71	0	0.0	0.0	0.0	0.0	0.0		
Blue Mountains	6.2.9	63	0	0.0	0.0	0.0	0.0	0.0		
Ozark Highlands	8.4.5	56	0	0.0	3.6	3.6	3.6	3.6		
Ouachita Mountains	8.4.8	42	0	0.0	0.0	0.0	0.0	0.0		
Mississippi Valley Loess Plains	8.3.6	41	0	0.0	9.8	9.8	14.6	14.6		
Strait of Georgia/Puget Lowland	7.1.7	38	0	0.0	0.0	0.0	0.0	0.0		
Western Allegheny Plateau	8.4.3	35	0	0.0	0.0	0.0	0.0	0.0		

	Sulfur only - ANC = 30 ueq/L									
	No. = 69					% Exce	edances	5		
Ecoregion			C	L≤0	2018	-2020	2014	-2016		
Name	#	No.	No.	%	All	CL>0	All	CL>0		
Southern Michigan/Northern Indiana Drift Plains	8.1.6	33	0	0.0	3.0	3.0	3.0	3.0		
Arkansas Valley	8.4.7	31	0	0.0	0.0	0.0	3.2	3.2		
Canadian Rockies	6.2.4	31	0	0.0	0.0	0.0	0.0	0.0		
Eastern Cascades Slopes and Foothills	6.2.8	27	0	0.0	0.0	0.0	3.7	3.7		
Cross Timbers	9.4.5	26	0	0.0	3.8	3.8	3.8	3.8		
Western Corn Belt Plains	9.2.3	26	0	0.0	0.0	0.0	0.0	0.0		
Arizona/New Mexico Mountains	13.1.1	25	0	0.0	0.0	0.0	0.0	0.0		
Willamette Valley	7.1.9	24	0	0.0	0.0	0.0	0.0	0.0		
Boston Mountains	8.4.6	23	0	0.0	0.0	0.0	0.0	0.0		
Southern and Baja California Pine-Oak Mountains	11.1.3	22	1	4.5	4.5	0.0	4.5	0.0		
Central Irregular Plains	9.2.4	21	0	0.0	0.0	0.0	4.8	4.8		
California Coastal Sage, Chaparral, and Oak Woodlands	11.1.1	21	0	0.0	0.0	0.0	0.0	0.0		
Northern Basin and Range	10.1.3	20	0	0.0	0.0	0.0	0.0	0.0		
Mississippi Alluvial Plain	8.5.2	19	1	5.3	5.3	0.0	5.3	0.0		
Interior River Valleys and Hills	8.3.2	18	0	0.0	0.0	0.0	5.6	5.6		
Western Gulf Coastal Plain	9.5.1	16	0	0.0	0.0	0.0	0.0	0.0		
Central Basin and Range	10.1.5	16	0	0.0	0.0	0.0	0.0	0.0		
Driftless Area	8.1.5	15	0	0.0	0.0	0.0	0.0	0.0		
Erie Drift Plain	8.1.10	14	0	0.0	0.0	0.0	7.1	7.1		
Eastern Corn Belt Plains	8.2.4	14	0	0.0	0.0	0.0	0.0	0.0		
East Central Texas Plains	8.3.8	10	1	10.0	10.0	0.0	10.0	0.0		
Southeastern Wisconsin Till Plains	8.2.1	10	0	0.0	0.0	0.0	0.0	0.0		
Flint Hills	9.4.4	7	1	14.3	28.6	14.3	28.6	14.3		
Central Great Plains	9.4.2	5	0	0.0	0.0	0.0	0.0	0.0		
Wyoming Basin	10.1.4	3	0	0.0	0.0	0.0	0.0	0.0		
Texas Blackland Prairies	9.4.7	3	0	0.0	0.0	0.0	0.0	0.0		
Northern Minnesota Wetlands	5.2.2	2	0	0.0	0.0	0.0	50.0	50.0		
Snake River Plain	10.1.8	2	0	0.0	0.0	0.0	0.0	0.0		
Central California Valley	11.1.2	2	0	0.0	0.0	0.0	0.0	0.0		
Columbia Plateau	10.1.2	2	0	0.0	0.0	0.0	0.0	0.0		
Central Corn Belt Plains	8.2.3	2	0	0.0	0.0	0.0	0.0	0.0		
Northwestern Glaciated Plains	9.3.1	2	0	0.0	0.0	0.0	0.0	0.0		
Colorado Plateaus	10.1.6	1	0	0.0	0.0	0.0	0.0	0.0		

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Table 5A-20.Percent Ecoregion Exceedances of aquatic CLs for Sulfur only by ANC<br/>threshold of 30 µeq/L for deposition years of 2010-12, 2006-08 and 2001-03.

	Sulfur only - ANC = 20 ueq/L									
	No. = 69				_		% Exc	eedance	es	
Ecoregion	1		CL	.≤0	2010	-2012	2006	-2008	200	1-2003
Name	#	No.	No.	%	All	CL>0	All	CL>0	All	CL>0
Northern Appalachian and	5.3.1	2851	11	0.4	4.5	4.1	14.9	14.5	19.9	19.5
Rua Didao	011	1072	2	0.2	61	5.0	20.2	20.0	12 7	12 5
Dides and Valley	0.4.4	1972	3 0	0.2	0.1	0.9	32.3	32.2	43.7 20 F	43.3
Ridge and Valley	0.4.1	1292	2 1	0.2	9.0	9.0	29.3	29.2	30.0	30.3
Northeastern Coastel Zana	0.2.1	039	1	0.1	2.9	2.7	0.0	0.7	10.0	15.9
Northeastern Coastal Zone	0.1./	505		0.2	Z.I	1.9	0.3	0.1	10.1	9.9
Middle Decline	0.3.4	508	0	0.0	1.0	1.0	11.0	11.0	10.1	10.1
Mildale Rockies	0.2.10	496	0	0.0	0.8	0.8	0.8	0.8	0.8	0.8
Acadian Plains and Hills	0.1.0	494	2	0.4	4.7	4.3	10.5	10.1	10.1	9.7
Southeastern Plains	8.3.5	390	3	0.8	9.5	8.7	20.5	19.7	24.1	23.3
Central Appalachians	8.4.2	372	4	1.1	15.9	14.8	44.4	43.3	53.8	52.7
Southern Rockies	6.2.14	372	1	0.3	2.2	1.9	2.7	2.4	2.7	2.4
Sierra Nevada	6.2.12	353	11	3.1	5.1	2.0	5.1	2.0	6.2	3.1
Atlantic Coastal Pine Barrens	8.5.4	234	1	0.4	23.9	23.5	45.3	44.9	53.8	53.4
Northern Piedmont	8.3.1	231	0	0.0	1.7	1.7	6.5	6.5	7.8	7.8
North Central Appalachians	5.3.3	216	0	0.0	5.6	5.6	24.1	24.1	31.5	31.5
Northern Allegheny Plateau	8.1.3	199	0	0.0	2.0	2.0	7.5	7.5	9.5	9.5
Idaho Batholith	6.2.15	188	1	0.5	0.5	0.0	0.5	0.0	0.5	0.0
Cascades	6.2.7	179	4	2.2	3.9	1.7	3.9	1.7	3.9	1.7
North Cascades	6.2.5	162	1	0.6	0.6	0.0	0.6	0.0	0.6	0.0
South Central Plains	8.3.7	153	2	1.3	13.7	12.4	19.0	17.6	20.3	19.0
Southern Coastal Plain	8.5.3	142	1	0.7	28.9	28.2	35.2	34.5	49.3	48.6
Southwestern Appalachians	8.4.9	117	0	0.0	0.0	0.0	25.6	25.6	41.0	41.0
Coast Range	7.1.8	115	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Middle Atlantic Coastal Plain	8.5.1	105	1	1.0	5.7	4.8	20.0	19.0	24.8	23.8
Wasatch and Uinta Mountains	6.2.13	96	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
North Central Hardwood Forests	8.1.4	94	0	0.0	0.0	0.0	0.0	0.0	3.2	3.2
Columbia Mountains/Northern Rockies	6.2.3	86	1	1.2	3.5	2.3	3.5	2.3	3.5	2.3
Eastern Great Lakes Lowlands	8.1.1	83	1	1.2	1.2	0.0	2.4	1.2	6.0	4.8
Klamath Mountains	6.2.11	81	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Interior Plateau	8.3.3	71	0	0.0	5.6	5.6	8.5	8.5	12.7	12.7
Blue Mountains	6.2.9	63	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Ozark Highlands	8.4.5	56	0	0.0	3.6	3.6	3.6	3.6	3.6	3.6
Ouachita Mountains	8.4.8	42	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Mississippi Valley Loess Plains	8.3.6	41	0	0.0	12.2	12.2	17.1	17.1	19.5	19.5
Strait of Georgia/Puget Lowland	7.1.7	38	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0

	Sulfur only - ANC = 20 ueq/L									
	No. = 69				_		% Exc	eedance	es	
Ecoregion			CL	.≤0	2010	-2012	2006	-2008	200	1-2003
Name	#	No.	No.	%	All	CL>0	All	CL>0	All	CL>0
Western Allegheny Plateau	8.4.3	35	0	0.0	2.9	2.9	20.0	20.0	28.6	28.6
Southern Michigan/Northern Indiana Drift Plains	8.1.6	33	0	0.0	6.1	6.1	15.2	15.2	21.2	21.2
Arkansas Valley	8.4.7	31	0	0.0	3.2	3.2	3.2	3.2	3.2	3.2
Canadian Rockies	6.2.4	31	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Eastern Cascades Slopes and Foothills	6.2.8	27	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Cross Timbers	9.4.5	26	0	0.0	0.0	0.0	3.8	3.8	7.7	7.7
Western Corn Belt Plains	9.2.3	26	0	0.0	0.0	0.0	0.0	0.0	3.8	3.8
Arizona/New Mexico Mountains	13.1.1	25	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Willamette Valley	7.1.9	24	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Boston Mountains	8.4.6	23	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Southern and Baja California Pine-Oak Mountains	11.1.3	22	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Central Irregular Plains	9.2.4	21	0	0.0	0.0	0.0	4.8	4.8	4.8	4.8
California Coastal Sage, Chaparral, and Oak Woodlands	11.1.1	21	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Northern Basin and Range	10.1.3	20	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Mississippi Alluvial Plain	8.5.2	19	0	0.0	5.3	5.3	5.3	5.3	21.1	21.1
Interior River Valleys and Hills	8.3.2	18	0	0.0	5.6	5.6	16.7	16.7	22.2	22.2
Western Gulf Coastal Plain	9.5.1	16	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Central Basin and Range	10.1.5	16	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Driftless Area	8.1.5	15	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Erie Drift Plain	8.1.10	14	0	0.0	7.1	7.1	21.4	21.4	28.6	28.6
Eastern Corn Belt Plains	8.2.4	14	0	0.0	14.3	14.3	28.6	28.6	28.6	28.6
East Central Texas Plains	8.3.8	10	0	0.0	10.0	10.0	10.0	10.0	10.0	10.0
Southeastern Wisconsin Till Plains	8.2.1	10	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Flint Hills	9.4.4	7	0	0.0	28.6	28.6	28.6	28.6	28.6	28.6
Central Great Plains	9.4.2	5	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Wyoming Basin	10.1.4	3	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Texas Blackland Prairies	9.4.7	3	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Northern Minnesota Wetlands	5.2.2	2	0	0.0	50.0	50.0	50.0	50.0	50.0	50.0
Snake River Plain	10.1.8	2	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Central California Valley	11.1.2	2	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Columbia Plateau	10.1.2	2	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Central Corn Belt Plains	8.2.3	2	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Northwestern Glaciated Plains	9.3.1	2	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Colorado Plateaus	10.1.6	1	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0

#### 1 Table 5A-21. Percent Ecoregion Exceedances of aquatic CLs for Sulfur only by ANC threshold of 50 µeq/L for deposition years of 2018-20 and 2014-16.

	Sulfur only - ANC = 50 ueq/L									
	No. = 69					% Exce	edances			
Ecoregion			CL≤0		2018	-2020	2014	-2016		
Name	#	No.	No.	%	All	CL>0	All	CL>0		
Northern Appalachian and Atlantic	5.3.1	2851	153	5.4	9.7	4.3	11.9	6.6		
Maritime Highlands										
Blue Ridge	8.4.4	1972	103	5.2	12.7	7.5	15.1	9.8		
Ridge and Valley	8.4.1	1292	28	2.2	5.3	3.1	10.1	8.0		
Northern Lakes and Forests	5.2.1	839	11	1.3	4.9	3.6	9.3	8.0		
Northeastern Coastal Zone	8.1.7	565	9	1.6	2.8	1.2	3.4	1.8		
Piedmont	8.3.4	508	6	1.2	3.5	2.4	4.5	3.3		
Middle Rockies	6.2.10	496	16	3.2	4.4	1.2	5.0	1.8		
Acadian Plains and Hills	8.1.8	494	29	5.9	7.9	2.0	8.9	3.0		
Southeastern Plains	8.3.5	390	21	5.4	12.1	6.7	13.8	8.5		
Central Appalachians	8.4.2	372	22	5.9	11.6	5.6	19.6	13.7		
Southern Rockies	6.2.14	372	30	8.1	9.4	1.3	11.3	3.2		
Sierra Nevada	6.2.12	353	90	25.5	28.3	2.8	30.0	4.5		
Atlantic Coastal Pine Barrens	8.5.4	234	28	12.0	24.8	12.8	27.8	15.8		
Northern Piedmont	8.3.1	231	3	1.3	1.7	0.4	2.6	1.3		
North Central Appalachians	5.3.3	216	5	2.3	4.2	1.9	6.9	4.6		
Northern Allegheny Plateau	8.1.3	199	4	2.0	2.5	0.5	2.5	0.5		
Idaho Batholith	6.2.15	188	9	4.8	5.9	1.1	9.6	4.8		
Cascades	6.2.7	179	21	11.7	13.4	1.7	12.8	1.1		
North Cascades	6.2.5	162	4	2.5	3.7	1.2	3.1	0.6		
South Central Plains	8.3.7	153	8	5.2	19.0	13.7	19.6	14.4		
Southern Coastal Plain	8.5.3	142	12	8.5	29.6	21.1	33.1	24.6		
Southwestern Appalachians	8.4.9	117	0	0.0	0.0	0.0	0.9	0.9		
Coast Range	7.1.8	115	0	0.0	0.0	0.0	0.0	0.0		
Middle Atlantic Coastal Plain	8.5.1	105	3	2.9	5.7	2.9	5.7	2.9		
Wasatch and Uinta Mountains	6.2.13	96	1	1.0	1.0	0.0	1.0	0.0		
North Central Hardwood Forests	8.1.4	94	1	1.1	2.1	1.1	3.2	2.1		
Columbia Mountains/Northern Rockies	6.2.3	86	4	4.7	5.8	1.2	5.8	1.2		
Eastern Great Lakes Lowlands	8.1.1	83	1	1.2	2.4	1.2	2.4	1.2		
Klamath Mountains	6.2.11	81	0	0.0	0.0	0.0	0.0	0.0		
Interior Plateau	8.3.3	71	2	2.8	7.0	4.2	7.0	4.2		
Blue Mountains	6.2.9	63	0	0.0	0.0	0.0	1.6	1.6		
Ozark Highlands	8.4.5	56	1	1.8	3.6	1.8	3.6	1.8		
Ouachita Mountains	8.4.8	42	0	0.0	2.4	2.4	2.4	2.4		
Mississippi Valley Loess Plains	8.3.6	41	1	2.4	19.5	17.1	19.5	17.1		
Strait of Georgia/Puget Lowland	7.1.7	38	1	2.6	2.6	0.0	5.3	2.6		
Western Allegheny Plateau	8.4.3	35	0	0.0	0.0	0.0	2.9	2.9		

	Sulfur only - ANC = 50 ueq/L									
	No. = 69					% Exce	edances	6		
Ecoregion	I		CL≤0	)	2018	-2020	2014-2016			
Name	#	No.	No.	%	All	CL>0	All	CL>0		
Southern Michigan/Northern Indiana	8.1.6	33	1	3.0	6.1	3.0	9.1	6.1		
Drift Plains										
Arkansas Valley	8.4.7	31	0	0.0	6.5	6.5	3.2	3.2		
Canadian Rockies	6.2.4	31	1	3.2	3.2	0.0	3.2	0.0		
Eastern Cascades Slopes and Foothills	6.2.8	27	0	0.0	0.0	0.0	3.7	3.7		
Cross Timbers	9.4.5	26	2	7.7	11.5	3.8	11.5	3.8		
Western Corn Belt Plains	9.2.3	26	1	3.8	3.8	0.0	3.8	0.0		
Arizona/New Mexico Mountains	13.1.1	25	0	0.0	0.0	0.0	0.0	0.0		
Willamette Valley	7.1.9	24	0	0.0	0.0	0.0	0.0	0.0		
Boston Mountains	8.4.6	23	0	0.0	0.0	0.0	0.0	0.0		
Southern and Baja California Pine-Oak Mountains	11.1.3	22	1	4.5	4.5	0.0	9.1	4.5		
Central Irregular Plains	9.2.4	21	1	4.8	9.5	4.8	14.3	9.5		
California Coastal Sage, Chaparral, and Oak Woodlands	11.1.1	21	0	0.0	0.0	0.0	0.0	0.0		
Northern Basin and Range	10.1.3	20	1	5.0	5.0	0.0	5.0	0.0		
Mississippi Alluvial Plain	8.5.2	19	1	5.3	15.8	10.5	15.8	10.5		
Interior River Valleys and Hills	8.3.2	18	0	0.0	11.1	11.1	11.1	11.1		
Western Gulf Coastal Plain	9.5.1	16	0	0.0	0.0	0.0	0.0	0.0		
Central Basin and Range	10.1.5	16	0	0.0	0.0	0.0	0.0	0.0		
Driftless Area	8.1.5	15	0	0.0	0.0	0.0	0.0	0.0		
Erie Drift Plain	8.1.10	14	0	0.0	7.1	7.1	7.1	7.1		
Eastern Corn Belt Plains	8.2.4	14	0	0.0	14.3	14.3	14.3	14.3		
East Central Texas Plains	8.3.8	10	1	10.0	10.0	0.0	10.0	0.0		
Southeastern Wisconsin Till Plains	8.2.1	10	0	0.0	0.0	0.0	0.0	0.0		
Flint Hills	9.4.4	7	2	28.6	28.6	0.0	28.6	0.0		
Central Great Plains	9.4.2	5	0	0.0	0.0	0.0	0.0	0.0		
Wyoming Basin	10.1.4	3	0	0.0	0.0	0.0	0.0	0.0		
Texas Blackland Prairies	9.4.7	3	0	0.0	0.0	0.0	0.0	0.0		
Northern Minnesota Wetlands	5.2.2	2	0	0.0	50.0	50.0	50.0	50.0		
Snake River Plain	10.1.8	2	0	0.0	0.0	0.0	0.0	0.0		
Central California Valley	11.1.2	2	0	0.0	0.0	0.0	0.0	0.0		
Columbia Plateau	10.1.2	2	0	0.0	0.0	0.0	0.0	0.0		
Central Corn Belt Plains	8.2.3	2	0	0.0	0.0	0.0	0.0	0.0		
Northwestern Glaciated Plains	9.3.1	2	0	0.0	0.0	0.0	0.0	0.0		
Colorado Plateaus	10.1.6	1	0	0.0	0.0	0.0	0.0	0.0		

1Table 5A-22.Percent Ecoregion Exceedances of aquatic CLs for Sulfur only by ANC2threshold of 50 μeq/L for deposition years of 2010-12, 2006-08 and 2001-03.

	Sulfur only - ANC = 50 ueq/L									
	No. = 69 % Exceedances						s			
Ecoregion			CL≤0		2010-2012		2006-2008		2001-2003	
Name	#	No.	No.	%	All	CL>0	All	CL>0	All	CL>0
Northern Appalachian and Atlantic	5.3.1	2851	153	5	15.0	9.7	28.0	22.6	32.0	26.6
Maritime Highlands										
Blue Ridge	8.4.4	1972	103	5	26.6	21.4	55.5	50.3	63.1	57.9
Ridge and Valley	8.4.1	1292	28	2	17.3	15.2	40.3	38.2	48.5	46.3
Northern Lakes and Forests	5.2.1	839	11	1	13.6	12.3	20.0	18.7	26.7	25.4
Northeastern Coastal Zone	8.1.7	565	9	2	5.3	3.7	15.0	13.5	16.5	14.9
Piedmont	8.3.4	508	6	1	6.9	5.7	20.9	19.7	25.0	23.8
Middle Rockies	6.2.10	496	16	3	5.6	2.4	6.3	3.0	5.8	2.6
Acadian Plains and Hills	8.1.8	494	29	6	10.9	5.1	18.6	12.8	17.6	11.7
Southeastern Plains	8.3.5	390	21	5	16.2	10.8	25.4	20.0	29.5	24.1
Central Appalachians	8.4.2	372	22	6	33.6	27.7	55.1	49.2	63.2	57.3
Southern Rockies	6.2.14	372	30	8	12.1	4.0	12.9	4.8	12.9	4.8
Sierra Nevada	6.2.12	353	90	25	30.0	4.5	30.0	4.5	30.0	4.5
Atlantic Coastal Pine Barrens	8.5.4	234	28	12	33.8	21.8	53.0	41.0	61.1	49.1
Northern Piedmont	8.3.1	231	3	1	3.5	2.2	8.2	6.9	10.4	9.1
North Central Appalachians	5.3.3	216	5	2	14.4	12.0	34.3	31.9	43.1	40.7
Northern Allegheny Plateau	8.1.3	199	4	2	4.0	2.0	11.6	9.5	14.6	12.6
Idaho Batholith	6.2.15	188	9	5	10.1	5.3	9.6	4.8	7.4	2.7
Cascades	6.2.7	179	21	12	12.8	1.1	14.0	2.2	13.4	1.7
North Cascades	6.2.5	162	4	2	3.7	1.2	3.7	1.2	3.7	1.2
South Central Plains	8.3.7	153	8	5	19.6	14.4	24.2	19.0	24.8	19.6
Southern Coastal Plain	8.5.3	142	12	8	33.8	25.4	40.8	32.4	53.5	45.1
Southwestern Appalachians	8.4.9	117	0	0	2.6	2.6	39.3	39.3	53.8	53.8
Coast Range	7.1.8	115	0	0	0.0	0.0	0.0	0.0	0.0	0.0
Middle Atlantic Coastal Plain	8.5.1	105	3	3	9.5	6.7	26.7	23.8	29.5	26.7
Wasatch and Uinta Mountains	6.2.13	96	1	1	1.0	0.0	1.0	0.0	1.0	0.0
North Central Hardwood Forests	8.1.4	94	1	1	4.3	3.2	6.4	5.3	10.6	9.6
Columbia Mountains/Northern	6.2.3	86	4	5	5.8	1.2	5.8	1.2	5.8	1.2
Rockies										
Eastern Great Lakes Lowlands	8.1.1	83	1	1	2.4	1.2	6.0	4.8	6.0	4.8
Klamath Mountains	6.2.11	81	0	0	0.0	0.0	0.0	0.0	0.0	0.0
Interior Plateau	8.3.3	71	2	3	8.5	5.6	11.3	8.5	15.5	12.7
Blue Mountains	6.2.9	63	0	0	0.0	0.0	0.0	0.0	0.0	0.0
Ozark Highlands	8.4.5	56	1	2	5.4	3.6	5.4	3.6	5.4	3.6
Ouachita Mountains	8.4.8	42	0	0	7.1	7.1	11.9	11.9	11.9	11.9
Mississippi Valley Loess Plains	8.3.6	41	1	2	19.5	17.1	19.5	17.1	22.0	19.5
Strait of Georgia/Puget Lowland	7.1.7	38	1	3	2.6	0.0	2.6	0.0	5.3	2.6

	Sulfur only - ANC = 50 ueq/L									
	No. = 69 % Exceedances									
Ecoregion			CL≤0		2010-2012		2006-2008		2001-2003	
Name	#	No.	No.	%	All	CL>0	All	CL>0	All	CL>0
Western Allegheny Plateau	8.4.3	35	0	0	11.4	11.4	25.7	25.7	37.1	37.1
Southern Michigan/Northern Indiana Drift Plains	8.1.6	33	1	3	15.2	12.1	27.3	24.2	27.3	24.2
Arkansas Valley	8.4.7	31	0	0	6.5	6.5	6.5	6.5	9.7	9.7
Canadian Rockies	6.2.4	31	1	3	3.2	0.0	3.2	0.0	3.2	0.0
Eastern Cascades Slopes and Foothills	6.2.8	27	0	0	0.0	0.0	0.0	0.0	0.0	0.0
Cross Timbers	9.4.5	26	2	8	11.5	3.8	15.4	7.7	19.2	11.5
Western Corn Belt Plains	9.2.3	26	1	4	3.8	0.0	15.4	11.5	15.4	11.5
Arizona/New Mexico Mountains	13.1.1	25	0	0	0.0	0.0	0.0	0.0	0.0	0.0
Willamette Valley	7.1.9	24	0	0	0.0	0.0	0.0	0.0	0.0	0.0
Boston Mountains	8.4.6	23	0	0	4.3	4.3	8.7	8.7	8.7	8.7
Southern and Baja California Pine- Oak Mountains	11.1.3	22	1	5	9.1	4.5	9.1	4.5	9.1	4.5
Central Irregular Plains	9.2.4	21	1	5	14.3	9.5	14.3	9.5	14.3	9.5
California Coastal Sage, Chaparral, and Oak Woodlands	11.1.1	21	0	0	0.0	0.0	0.0	0.0	0.0	0.0
Northern Basin and Range	10.1.3	20	1	5	5.0	0.0	5.0	0.0	5.0	0.0
Mississippi Alluvial Plain	8.5.2	19	1	5	15.8	10.5	26.3	21.1	26.3	21.1
Interior River Valleys and Hills	8.3.2	18	0	0	11.1	11.1	22.2	22.2	22.2	22.2
Western Gulf Coastal Plain	9.5.1	16	0	0	0.0	0.0	0.0	0.0	0.0	0.0
Central Basin and Range	10.1.5	16	0	0	0.0	0.0	0.0	0.0	0.0	0.0
Driftless Area	8.1.5	15	0	0	0.0	0.0	0.0	0.0	0.0	0.0
Erie Drift Plain	8.1.10	14	0	0	14.3	14.3	28.6	28.6	35.7	35.7
Eastern Corn Belt Plains	8.2.4	14	0	0	28.6	28.6	28.6	28.6	28.6	28.6
East Central Texas Plains	8.3.8	10	1	10	10.0	0.0	20.0	10.0	20.0	10.0
Southeastern Wisconsin Till Plains	8.2.1	10	0	0	0.0	0.0	0.0	0.0	0.0	0.0
Flint Hills	9.4.4	7	2	29	28.6	0.0	28.6	0.0	28.6	0.0
Central Great Plains	9.4.2	5	0	0	0.0	0.0	0.0	0.0	0.0	0.0
Wyoming Basin	10.1.4	3	0	0	0.0	0.0	0.0	0.0	0.0	0.0
Texas Blackland Prairies	9.4.7	3	0	0	0.0	0.0	0.0	0.0	0.0	0.0
Northern Minnesota Wetlands	5.2.2	2	0	0	50.0	50.0	50.0	50.0	50.0	50.0
Snake River Plain	10.1.8	2	0	0	0.0	0.0	0.0	0.0	0.0	0.0
Central California Valley	11.1.2	2	0	0	0.0	0.0	0.0	0.0	0.0	0.0
Columbia Plateau	10.1.2	2	0	0	0.0	0.0	0.0	0.0	0.0	0.0
Central Corn Belt Plains	8.2.3	2	0	0	0.0	0.0	0.0	0.0	0.0	0.0
Northwestern Glaciated Plains	9.3.1	2	0	0	0.0	0.0	0.0	0.0	0.0	0.0
Colorado Plateaus	10.1.6	1	0	0	0.0	0.0	0.0	0.0	0.0	0.0

### Percent Ecoregion Exceedances of aquatic CLs for Sulfur only by ANC threshold of 50/20 µeq/L for deposition years of 2018-20 and 2014-16. 1 Table 5A-23.

	Sulfur only - ANC = 50/20 ueq/L									
	No. = 69		% Exceedances							
Ecoregion			CL≤0		2018-2020		2014-2016			
Name	#	No.	No.	%	All	CL>0	All	CL>0		
Northern Appalachian and Atlantic	5.3.1	2851	153	5.4	9.7	4.3	11.9	6.6		
Maritime Highlands										
Blue Ridge	8.4.4	1972	103	5.2	12.7	7.5	15.1	9.8		
Ridge and Valley	8.4.1	1292	28	2.2	5.3	3.1	10.1	8.0		
Northern Lakes and Forests	5.2.1	839	11	1.3	4.9	3.6	9.3	8.0		
Northeastern Coastal Zone	8.1.7	565	9	1.6	2.8	1.2	3.4	1.8		
Piedmont	8.3.4	508	6	1.2	3.5	2.4	4.5	3.3		
Middle Rockies	6.2.10	496	0	0.0	0.2	0.2	0.6	0.6		
Acadian Plains and Hills	8.1.8	494	29	5.9	7.9	2.0	8.9	3.0		
Southeastern Plains	8.3.5	390	21	5.4	12.1	6.7	13.8	8.5		
Central Appalachians	8.4.2	372	22	5.9	11.6	5.6	19.6	13.7		
Southern Rockies	6.2.14	372	1	0.3	0.5	0.3	1.9	1.6		
Sierra Nevada	6.2.12	353	11	3.1	4.2	1.1	5.9	2.8		
Atlantic Coastal Pine Barrens	8.5.4	234	28	12.0	24.8	12.8	27.8	15.8		
Northern Piedmont	8.3.1	231	3	1.3	1.7	0.4	2.6	1.3		
North Central Appalachians	5.3.3	216	5	2.3	4.2	1.9	6.9	4.6		
Northern Allegheny Plateau	8.1.3	199	4	2.0	2.5	0.5	2.5	0.5		
Idaho Batholith	6.2.15	188	1	0.5	0.5	0.0	0.5	0.0		
Cascades	6.2.7	179	4	2.2	3.9	1.7	5.0	2.8		
North Cascades	6.2.5	162	1	0.6	0.6	0.0	0.6	0.0		
South Central Plains	8.3.7	153	8	5.2	19.0	13.7	19.6	14.4		
Southern Coastal Plain	8.5.3	142	12	8.5	29.6	21.1	33.1	24.6		
Southwestern Appalachians	8.4.9	117	0	0.0	0.0	0.0	0.9	0.9		
Coast Range	7.1.8	115	0	0.0	0.0	0.0	0.0	0.0		
Middle Atlantic Coastal Plain	8.5.1	105	3	2.9	5.7	2.9	5.7	2.9		
Wasatch and Uinta Mountains	6.2.13	96	0	0.0	0.0	0.0	0.0	0.0		
North Central Hardwood Forests	8.1.4	94	1	1.1	2.1	1.1	3.2	2.1		
Columbia Mountains/Northern Rockies	6.2.3	86	1	1.2	1.2	0.0	2.3	1.2		
Eastern Great Lakes Lowlands	8.1.1	83	1	1.2	2.4	1.2	2.4	1.2		
Klamath Mountains	6.2.11	81	0	0.0	0.0	0.0	0.0	0.0		
Interior Plateau	8.3.3	71	2	2.8	7.0	4.2	7.0	4.2		
Blue Mountains	6.2.9	63	0	0.0	0.0	0.0	0.0	0.0		
Ozark Highlands	8.4.5	56	1	1.8	3.6	1.8	3.6	1.8		
Ouachita Mountains	8.4.8	42	0	0.0	2.4	2.4	2.4	2.4		
Mississippi Valley Loess Plains	8.3.6	41	1	2.4	19.5	17.1	19.5	17.1		
Strait of Georgia/Puget Lowland	7.1.7	38	0	0.0	0.0	0.0	0.0	0.0		
Western Allegheny Plateau	8.4.3	35	0	0.0	0.0	0.0	2.9	2.9		
	Sulfur only - ANC = 50/20 ueq/L									
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	No. = 69					% Exce	edances	5		
Ecoregion			C	L≤0	2018	-2020	2014-2016			
Name	#	No.	No.	%	All CL>0		All	CL>0		
Southern Michigan/Northern Indiana	8.1.6	33	1	3.0	6.1	3.0	9.1	6.1		
Drift Plains										
Arkansas Valley	8.4.7	31	0	0.0	6.5	6.5	3.2	3.2		
Canadian Rockies	6.2.4	31	0	0.0	0.0	0.0	0.0	0.0		
Eastern Cascades Slopes and Foothills	6.2.8	27	0	0.0	0.0	0.0	0.0	0.0		
Cross Timbers	9.4.5	26	2	7.7	11.5	3.8	11.5	3.8		
Western Corn Belt Plains	9.2.3	26	1	3.8	3.8	0.0	3.8	0.0		
Arizona/New Mexico Mountains	13.1.1	25	0	0.0	0.0	0.0	0.0	0.0		
Willamette Valley	7.1.9	24	0	0.0	0.0	0.0	0.0	0.0		
Boston Mountains	8.4.6	23	0	0.0	0.0	0.0	0.0	0.0		
Southern and Baja California Pine-Oak Mountains	11.1.3	22	0	0.0	0.0	0.0	4.5	4.5		
Central Irregular Plains	9.2.4	21	1	4.8	9.5	4.8	14.3	9.5		
California Coastal Sage, Chaparral, and Oak Woodlands	11.1.1	21	0	0.0	0.0	0.0	0.0	0.0		
Northern Basin and Range	10.1.3	20	0	0.0	0.0	0.0	0.0	0.0		
Mississippi Alluvial Plain	8.5.2	19	1	5.3	15.8	10.5	15.8	10.5		
Interior River Valleys and Hills	8.3.2	18	0	0.0	11.1	11.1	11.1	11.1		
Western Gulf Coastal Plain	9.5.1	16	0	0.0	0.0	0.0	0.0	0.0		
Central Basin and Range	10.1.5	16	0	0.0	0.0	0.0	0.0	0.0		
Driftless Area	8.1.5	15	0	0.0	0.0	0.0	0.0	0.0		
Erie Drift Plain	8.1.10	14	0	0.0	7.1	7.1	7.1	7.1		
Eastern Corn Belt Plains	8.2.4	14	0	0.0	14.3	14.3	14.3	14.3		
East Central Texas Plains	8.3.8	10	1	10.0	10.0	0.0	10.0	0.0		
Southeastern Wisconsin Till Plains	8.2.1	10	0	0.0	0.0	0.0	0.0	0.0		
Flint Hills	9.4.4	7	2	28.6	28.6	0.0	28.6	0.0		
Central Great Plains	9.4.2	5	0	0.0	0.0	0.0	0.0	0.0		
Wyoming Basin	10.1.4	3	0	0.0	0.0	0.0	0.0	0.0		
Texas Blackland Prairies	9.4.7	3	0	0.0	0.0	0.0	0.0	0.0		
Northern Minnesota Wetlands	5.2.2	2	0	0.0	50.0	50.0	50.0	50.0		
Snake River Plain	10.1.8	2	0	0.0	0.0	0.0	0.0	0.0		
Central California Valley	11.1.2	2	0	0.0	0.0	0.0	0.0	0.0		
Columbia Plateau	10.1.2	2	0	0.0	0.0	0.0	0.0	0.0		
Central Corn Belt Plains	8.2.3	2	0	0.0	0.0	0.0	0.0	0.0		
Northwestern Glaciated Plains	9.3.1	2	0	0.0	0.0	0.0	0.0	0.0		
Colorado Plateaus	10.1.6	1	0	0.0	0.0	0.0	0.0	0.0		

### Table 5A-24.Percent Ecoregion Exceedances of aquatic CLs for Sulfur only by ANC<br/>threshold of 50/20 μeq/L for deposition years of 2010-12, 2006-08 and 2001-<br/>03.

	Sulfur only - ANC = 50/20 ueq/L									
	No. = 69						% Exc	eedance	es	
Ecoregion			CI	_≤0	2010	-2012	2006	-2008	2001-2003	
Name	#	No.	No.	%	All	CL>0	All	CL>0	All	CL>0
Northern Appalachian and Atlantic Maritime Highlands	5.3.1	2851	153	5.4	15.0	9.7	28.0	22.6	32.0	26.6
Blue Ridge	8.4.4	1972	103	5.2	26.6	21.4	55.5	50.3	63.1	57.9
Ridge and Valley	8.4.1	1292	28	2.2	17.3	15.2	40.3	38.2	48.5	46.3
Northern Lakes and Forests	5.2.1	839	11	1.3	13.6	12.3	20.0	18.7	26.7	25.4
Northeastern Coastal Zone	8.1.7	565	9	1.6	5.3	3.7	15.0	13.5	16.5	14.9
Piedmont	8.3.4	508	6	1.2	6.9	5.7	20.9	19.7	25.0	23.8
Middle Rockies	6.2.10	496	0	0.0	0.8	0.8	0.8	0.8	0.8	0.8
Acadian Plains and Hills	8.1.8	494	29	5.9	10.9	5.1	18.6	12.8	17.6	11.7
Southeastern Plains	8.3.5	390	21	5.4	16.2	10.8	25.4	20.0	29.5	24.1
Central Appalachians	8.4.2	372	22	5.9	33.6	27.7	55.1	49.2	63.2	57.3
Southern Rockies	6.2.14	372	1	0.3	2.2	1.9	2.7	2.4	2.7	2.4
Sierra Nevada	6.2.12	353	11	3.1	5.1	2.0	5.1	2.0	6.2	3.1
Atlantic Coastal Pine Barrens	8.5.4	234	28	12.0	33.8	21.8	53.0	41.0	61.1	49.1
Northern Piedmont	8.3.1	231	3	1.3	3.5	2.2	8.2	6.9	10.4	9.1
North Central Appalachians	5.3.3	216	5	2.3	14.4	12.0	34.3	31.9	43.1	40.7
Northern Allegheny Plateau	8.1.3	199	4	2.0	4.0	2.0	11.6	9.5	14.6	12.6
Idaho Batholith	6.2.15	188	1	0.5	0.5	0.0	0.5	0.0	0.5	0.0
Cascades	6.2.7	179	4	2.2	3.9	1.7	3.9	1.7	3.9	1.7
North Cascades	6.2.5	162	1	0.6	0.6	0.0	0.6	0.0	0.6	0.0
South Central Plains	8.3.7	153	8	5.2	19.6	14.4	24.2	19.0	24.8	19.6
Southern Coastal Plain	8.5.3	142	12	8.5	33.8	25.4	40.8	32.4	53.5	45.1
Southwestern Appalachians	8.4.9	117	0	0.0	2.6	2.6	39.3	39.3	53.8	53.8
Coast Range	7.1.8	115	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Middle Atlantic Coastal Plain	8.5.1	105	3	2.9	9.5	6.7	26.7	23.8	29.5	26.7
Wasatch and Uinta Mountains	6.2.13	96	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
North Central Hardwood Forests	8.1.4	94	1	1.1	4.3	3.2	6.4	5.3	10.6	9.6
Columbia Mountains/Northern Rockies	6.2.3	86	1	1.2	3.5	2.3	3.5	2.3	3.5	2.3
Eastern Great Lakes Lowlands	8.1.1	83	1	1.2	2.4	1.2	6.0	4.8	6.0	4.8
Klamath Mountains	6.2.11	81	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Interior Plateau	8.3.3	71	2	2.8	8.5	5.6	11.3	8.5	15.5	12.7
Blue Mountains	6.2.9	63	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Ozark Highlands	8.4.5	56	1	1.8	5.4	3.6	5.4	3.6	5.4	3.6
Ouachita Mountains	8.4.8	42	0	0.0	7.1	7.1	11.9	11.9	11.9	11.9
Mississippi Valley Loess Plains	8.3.6	41	1	2.4	19.5	17.1	19.5	17.1	22.0	19.5

	Sulfur only - ANC = 50/20 ueq/L									
	No. = 69				_		% Exc	eedance	es	
Ecoregion	1		CI	_≤0	2010	-2012	2006	-2008	200	1-2003
Name	#	No.	No.	%	All	CL>0	All	CL>0	All	CL>0
Strait of Georgia/Puget Lowland	7.1.7	38	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Western Allegheny Plateau	8.4.3	35	0	0.0	11.4	11.4	25.7	25.7	37.1	37.1
Southern Michigan/Northern	8.1.6	33	1	3.0	15.2	12.1	27.3	24.2	27.3	24.2
Indiana Drift Plains										
Arkansas Valley	8.4.7	31	0	0.0	6.5	6.5	6.5	6.5	9.7	9.7
Canadian Rockies	6.2.4	31	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Eastern Cascades Slopes and	6.2.8	27	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Foothills	0.4.5		-		44.5		45.4		40.0	44 5
	9.4.5	26	2	1.1	11.5	3.8	15.4	1.1	19.2	11.5
Western Corn Belt Plains	9.2.3	26	1	3.8	3.8	0.0	15.4	11.5	15.4	11.5
Arizona/New Mexico Mountains	13.1.1	25	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Willamette Valley	7.1.9	24	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Boston Mountains	8.4.6	23	0	0.0	4.3	4.3	8.7	8.7	8.7	8.7
Southern and Baja California Pine-Oak Mountains	11.1.3	22	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Central Irregular Plains	9.2.4	21	1	4.8	14.3	9.5	14.3	9.5	14.3	9.5
California Coastal Sage, Chaparral, and Oak Woodlands	11.1.1	21	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Northern Basin and Range	10.1.3	20	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Mississippi Alluvial Plain	8.5.2	19	1	5.3	15.8	10.5	26.3	21.1	26.3	21.1
Interior River Valleys and Hills	8.3.2	18	0	0.0	11.1	11.1	22.2	22.2	22.2	22.2
Western Gulf Coastal Plain	9.5.1	16	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Central Basin and Range	10.1.5	16	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Driftless Area	8.1.5	15	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Erie Drift Plain	8.1.10	14	0	0.0	14.3	14.3	28.6	28.6	35.7	35.7
Eastern Corn Belt Plains	8.2.4	14	0	0.0	28.6	28.6	28.6	28.6	28.6	28.6
East Central Texas Plains	8.3.8	10	1	10.0	10.0	0.0	20.0	10.0	20.0	10.0
Southeastern Wisconsin Till Plains	8.2.1	10	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Flint Hills	9.4.4	7	2	28.6	28.6	0.0	28.6	0.0	28.6	0.0
Central Great Plains	9.4.2	5	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Wyoming Basin	10.1.4	3	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Texas Blackland Prairies	9.4.7	3	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Northern Minnesota Wetlands	5.2.2	2	0	0.0	50.0	50.0	50.0	50.0	50.0	50.0
Snake River Plain	10.1.8	2	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Central California Valley	11.1.2	2	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Columbia Plateau	10.1.2	2	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Central Corn Belt Plains	8.2.3	2	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Northwestern Glaciated Plains	9.3.1	2	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Colorado Plateaus	10.1.6	1	0	0.0	0.0	0.0	0.0	0.0	0.0	0.0





2014 - 2016 Sulfur Deposition Ecoregion Exceedances



deposition from 2018-20 (top) and 2014-16 (bottom) for an ANC threshold of 20 μeq/L. Ecoregions with less than 50 critical loads are shaded and ecoregions without any values are blank. The Southern Coastal Plan (8.5.3) and Atlantic Coastal Pine Barrens (8.5.4) ecoregions are cross hatched to indicate natural high level of acidity.

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2010 - 2012 Sulfur Deposition Ecoregion Exceedances

2006 - 2008 Sulfur Deposition Ecoregion Exceedances



deposition from 2010-12 (top) and 2006-08 (bottom) for an ANC threshold
of 20 μeq/L. Ecoregions with less than 50 critical loads are shaded and
ecoregions without any values are blank. The Southern Coastal Plan
(8.5.3) and Atlantic Coastal Pine Barrens (8.5.4) ecoregions are cross
hatched to indicate natural high level of acidity.



### 2001 - 2003 Sulfur Deposition Ecoregion Exceedances

Figure 5A-34. Aggregated percent ecoregion critical load exceedances for S only deposition from 2001-02 for an ANC threshold of 20 μeq/L. Ecoregions with less than 50 critical loads are shaded and ecoregions without any values are blank. The Southern Coastal Plan (8.5.3) and Atlantic Coastal Pine Barrens (8.5.4) ecoregions are cross hatched to indicate natural high level of acidity.



2018 - 2020 Sulfur Deposition Ecoregion Exceedances

2014 - 2016 Sulfur Deposition Ecoregion Exceedances



ecoregions without any values are blank. The Southern Coastal Plan (8.5.3) and Atlantic Coastal Pine Barrens (8.5.4) ecoregions are cross hatched to indicate natural high level of acidity.

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2010 - 2012 Sulfur Deposition Ecoregion Exceedances

2006 - 2008 Sulfur Deposition Ecoregion Exceedances



ecoregions without any values are blank. The Southern Coastal Plan (8.5.3) and Atlantic Coastal Pine Barrens (8.5.4) ecoregions are cross 7 hatched to indicate natural high level of acidity.

### 2001 - 2003 Sulfur Deposition Ecoregion Exceedances



A-37. Aggregated percent ecoregion critical load exceedances for S only deposition from 2001-03 for an ANC threshold of 30 μeq/L. Ecoregions with less than 50 critical loads are shaded and ecoregions without any values are blank. The Southern Coastal Plan (8.5.3) and Atlantic Coastal Pine Barrens (8.5.4) ecoregions are cross hatched to indicate natural high level of acidity.



2018 - 2020 Sulfur Deposition Ecoregion Exceedances

2014 - 2016 Sulfur Deposition Ecoregion Exceedances



Figure 5A-38. Aggregated percent ecoregion critical load exceedances for S only deposition from 2018-20 (top) and 2014-16 (bottom) for an ANC threshold of 50 μeq/L. Ecoregions with less than 50 critical loads are shaded and ecoregions without any values are blank. The Southern Coastal Plan (8.5.3) and Atlantic Coastal Pine Barrens (8.5.4) ecoregions are cross hatched to indicate natural high level of acidity.



2010 - 2012 Sulfur Deposition Ecoregion Exceedances

2006 - 2008 Sulfur Deposition Ecoregion Exceedances



Figure 5A-39. Aggregated percent ecoregion critical load exceedances for S only deposition from 2010-12 (top) and 2006-08 (bottom) for an ANC threshold of 50 μeq/L. Ecoregions with less than 50 critical loads are shaded and ecoregions without any values are blank. The Southern Coastal Plan (8.5.3) and Atlantic Coastal Pine Barrens (8.5.4) ecoregions are cross hatched to indicate natural high level of acidity.

### 2001 - 2003 Sulfur Deposition Ecoregion Exceedances



gure 5A-40. Aggregated percent ecoregion critical load exceedances for S only deposition from 2001-03 for an ANC threshold of 50 µeq/L. Ecoregions with less than 50 critical loads are shaded and ecoregions without any values are blank. The Southern Coastal Plan (8.5.3) and Atlantic Coastal Pine Barrens (8.5.4) ecoregions are cross hatched to indicate natural high level of acidity.



2018 - 2020 Sulfur Deposition Ecoregion Exceedances

2014 - 2016 Sulfur Deposition Ecoregion Exceedances



deposition from 2018-20 (top) and 2014-16 (bottom) for an ANC threshold of 50 µeq/L for East and 20 µeq/L for the West. Ecoregions with less than 50 critical loads are shaded and ecoregions without any values are blank. The Southern Coastal Plan (8.5.3) and Atlantic Coastal Pine Barrens (8.5.4) 7 ecoregions are cross hatched to indicate natural high level of acidity.



2010 - 2012 Sulfur Deposition Ecoregion Exceedances

2006 - 2008 Sulfur Deposition Ecoregion Exceedances



2	Figure 5A-42.	Aggregated percent ecoregion critical load exceedances for S only
3		deposition from 2010-12 (top) and 2006-08 (bottom) for an ANC threshold
4		of 50 µeq/L for East and 20 µeq/L for the West. Ecoregions with less than
5		50 critical loads are shaded and ecoregions without any values are blank.
6		The Southern Coastal Plan (8.5.3) and Atlantic Coastal Pine Barrens (8.5.4)
7		ecoregions are cross hatched to indicate natural high level of acidity.



### 2001 - 2003 Sulfur Deposition Ecoregion Exceedances

Figure 5A-43. Aggregated percent ecoregion critical load exceedances for S only
deposition from 2001-03 for an ANC threshold of 50 µeq/L for East and 20
µeq/L for the West. Ecoregions with less than 50 critical loads are shaded
and ecoregions without any values are blank. The Southern Coastal Plan
(8.5.3) and Atlantic Coastal Pine Barrens (8.5.4) ecoregions are cross
hatched to indicate natural high level of acidity.

#### 5A.2.2.2 Ecoregion Summary – Percent Exceedances as a Function of Total S deposition

10 Ecoregions across the 5 deposition periods (2001-03, 2006-08, 2010-12, 2014-06, 2018-11 20) were summarized by the number of ecoregions with percent of EX over 10, 15, 20, 25, and 12 30% by total S deposition (e.g., 10% of all the CLs in the ecoregion EX). Ecoregions included in this analysis contain at least 50 CLs. A total of 25 acid sensitive ecoregions across the CONUS 13 14 with 18 and 7 ecoregions in the eastern and western U.S. have greater than 50 CLs. Table 5A-25 15 summarizes the min, max, and median total S deposition for ecoregions included in this analysis 16 with 50 or more CLs and positive exceedances. Deposition levels were summarized for the 5 17 deposition periods and 3 CL thresholds (20, 30, and 50  $\mu$ eq/L) for the eastern and western U.S. 18 separate and combined. Deposition for ecoregions in the eastern U.S. had a median value of 11.0 19 Kg S/ha-yr in 2001-03 and 1.9 Kg S/ha-yr in 2018-20. Total S deposition for ecoregions in the

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1 western U.S. was lower from a median of 1.14 Kg S/ha-yr in 2001-03 to 0.84 Kg S/ha-yr in

2 20180-20.

3 This summary is intended to look at the percent exceedances as a function of total S 4 deposition. For example, at 2 Kg S/ha-yr across all ecoregions and deposition periods that are no 5 ecoregions that have >10% EX for ANC threshold 50  $\mu$ eq/L (Tables 5A-26). The lowest 6 deposition level with EX>10% was at 3 Kg S/ha-yr, which had only 1 ecoregion. However, at 10 7 Kg S/ha-yr, there are 22 ecoregions across all deposition periods with >10% EX and 1 with 8 >30% EX. At 6, 10, 15 Kg S/ha-yr, there were 13, 22, 33 ecoregions with EX>10% and 2, 6, 14 9 ecoregions with EX>20%. Ecoregions with the most severe exceedances (>30%) started at 10 10 Kg S/ha-yr with 1 ecoregion and had 7 ecoregions with EX>30% at 10 Kg S/ha-yr. This was 11 done for ANC thresholds 20, 30, and 50 µeq/L for the eastern U.S., 20 µeq/L western U.S., and 12 combined 50/20, 30/20, and 20 µeg/L for both eastern and western U.S. Results are summarized 13 in Tables 5A-26, 5A-28, 5A-30, 5A-32, 5A-34, 5A-36, and 5A-38. 14 Cumulative percent of ecoregions across the 5 deposition periods were also determined 15 and graphed. This shows the percent of ecoregions as a function of deposition. For example, 16 for ANC of 50 µeq/L and for the eastern U.S, 100% of ecoregions and time periods have no 17 exceedances > 10% at 2 Kg S/ha-yr while at 19 Kg S/ha-yr 60% or of ecoregions and time periods have no EX > 10% (e.g 40% have > 10% EX) (Tables 5A-26, Figure 5A-44). Results 18 19 for the other ANC thresholds are summarized in Tables 5A-28, 5A-30, 5A-32, 5A-34, 5A-36, 20 5A-38. Cumulative results are graphed in Figure 5A-45 to 5A-50. Figure 5A-51 summarized 21 Total S deposition (Kg S/Ha-yr) as a function of percent of waterbodies exceeding the CLs for 22 2018-20 and 2014-16 for thresholds ANC = 20, 30, and 50  $\mu$ eq/L for positive CLS (CL>0). EX 23 >10% fall between 4-5 Kg S/ha-yr.

Minimum, maximum, and median S deposition for ecoregions with at least 1 Table 5A-25. 50 critical loads and with ecoregions with exceedances for the five deposition periods and three ANC targets. Deposition values were determined by a zonal statistic for each ecoregion. 4

	Median	Sulfur Dep	osition (Kg	S/ha-yr)	
	2001-03	2006-08	2010-12	2014-16	2018-20
	AI	VC Target of	<sup>2</sup> 0 µeq/L fo	r the East U	.S.
Min	4.3	3.2	2.4	1.6	1.2
Max	18.1	15.1	7.2	4.7	3.6
Median	11.0	9.3	4.6	3.0	1.9
	AI	VC Target of	<sup>r</sup> 30 µeq/L fo	r the East U	.S.
Min	4.3	3.2	2.4	1.6	1.2
Max	18.1	15.1	7.2	4.7	3.6
Median	11.0	9.0	4.6	2.8	1.9
	AI	VC Target of	$^{\circ}$ 50 $\mu$ eq/L fo	r the East U	.S.
Min	4.3	3.2	2.4	1.6	1.2
Max	18.1	15.1	7.2	4.7	3.6
Median	11.0	9.0	4.5	3.0	1.9
	ANC Targe	et of 20 µeq	L for the We	st U.S.	
Min	0.90	0.98	0.83	0.79	0.54
Max	1.69	1.66	1.41	1.51	1.24
Median	1.14	1.14	0.93	0.86	0.84
	ANC Targe	et of 20 µeq	/L for the Ea	st/West U.S.	
Min	0.90	0.98	0.83	0.79	0.54
Max	18.08	15.05	7.24	4.70	3.64
Median	9.57	8.24	4.34	2.61	1.51
	ANC Targe	et of 30/20 µ	eq/L for the	East/West L	J.S.
Min	0.90	0.98	0.83	0.79	0.54
Max	18.08	15.05	7.24	4.70	3.64
Median	9.57	8.05	4.34	2.50	1.87
	ANC Targe	et of 50/20 µ	eq/L for the	East/West L	J.S.
Min	0.90	0.98	0.83	0.79	0.54
Max	18.08	15.05	7.24	4.70	3.64
Median	9.57	8.05	4.34	2.62	1.87

#### 1 Table 5A-26. Number of ecoregion-time period combinations with more than 10, 15, 20, 2 3 25 and 30% of waterbodies exceeding their CLs for three ANC target of 50 µeq/L. Includes 18 ecoregions in the eastern U.S.

Total Sulfur Deposition	Number of Ecoregion with Percent of Exceedances Across the 5 deposition Periods						
Kg S/ha-yr	10%	15%	20%	25%	30%		
2	0	0	0	0	0		
3	1	0	0	0	0		
4	3	1	0	0	0		
5	9	3	2	1	0		
6	13	4	2	1	0		
7	14	5	3	1	0		
8	18	9	5	3	0		
9	19	9	5	3	0		
10	22	11	6	4	1		
11	24	13	7	4	1		
12	27	15	9	6	3		
13	28	16	10	6	3		
14	31	18	12	8	5		
15	33	20	14	10	7		
16	34	21	15	11	8		
17	34	21	15	11	8		
18	36	23	17	13	10		
19	36	23	17	13	10		

1Table 5A-27.Cumulative percentage of ecoregion-time period combinations with more2than 10, 15, 20, 25, and 30% of waterbodies per ecoregion meeting their3CLs for the ANC target of 50 μeq/L as a function of total S deposition4across all 5 deposition periods (2001-03, 2006-08, 2010-12, 2014-06, 2018-520). 100% indicates there were no ecoregions that had percent exceedances6above specified value. For the eastern U.S. (See Table 5A-27 for data).

Total Sulfur Deposition	Percent of Exceedances Across the 5 deposition Periods							
Kg S/ha-yr	10%	15%	20%	25%	30%			
2	100%	100%	100%	100%	100%			
3	97%	100%	100%	100%	100%			
4	93%	98%	100%	100%	100%			
5	82%	94%	96%	98%	100%			
6	78%	93%	97%	98%	100%			
7	78%	92%	95%	98%	100%			
8	73%	87%	93%	96%	100%			
9	72%	87%	93%	96%	100%			
10	70%	85%	92%	95%	99%			
11	68%	83%	91%	95%	99%			
12	66%	81%	89%	92%	96%			
13	65%	80%	88%	93%	96%			
14	63%	79%	86%	90%	94%			
15	62%	77%	84%	88%	92%			
16	61%	76%	83%	88%	91%			
17	61%	76%	83%	88%	91%			
18	60%	74%	81%	86%	89%			



across all 5 deposition periods (2001-03, 2006-08, 2010-12, 2014-06, 2018-

above >10, >15, >20, >25, >30% for that deposition level. Critical load

Table 5A-27 for data).

20). 100% indicates there were no ecoregion that had percent exceedances

exceedances based on ANC threshold of 50 µeq/L for the eastern U.S. (See

1Table 5A-28.Number of ecoregions with percent of exceedances of >10, >15, >20, >25,2>30% as a function of total S deposition across all 5 deposition periods3(2001-03, 2006-08, 2010-12, 2014-06, 2018-20). Includes 18 ecoregions in4the eastern U.S. and Critical load exceedances are based on an ANC5threshold of 30 μeq/L.

Total Sulfur	Number of Ecoregion with Percent of Exceedances Across the 5 deposition Periods						
Kg S/ha-yr	10%	15%	20%	25%	30%		
2	0	0	0	0	0		
3	0	0	0	0	0		
4	2	0	0	0	0		
5	4	1	0	0	0		
6	7	1	0	0	0		
7	8	2	0	0	0		
8	12	6	1	0	0		
9	13	6	1	0	0		
10	16	8	2	1	1		
11	18	9	3	1	1		
12	21	11	5	3	3		
13	22	12	5	3	3		
14	25	14	7	5	4		
15	27	16	9	7	6		
16	28	17	10	8	7		
17	28	17	10	8	7		
18	30	19	12	10	9		
19	30	19	12	10	9		

# 1Table 5A-29.Cumulative percent of waterbodies in ecoregions meeting the target ANC2values as a function of total S deposition across all 5 deposition periods3(2001-03, 2006-08, 2010-12, 2014-06, 2018-20).100% indicates there were4no ecoregions that had percent exceedances above >10, >15, >20, >25,5>30% for a given deposition level. Critical load exceedances based on ANC6threshold of 30 μeq/L for the eastern U.S. (See Table 5A-27 for data).

Total Sulfur Deposition	Number of Ecoregion with Percent of Exceedances Across the 5 deposition Periods						
Kg <b>S/ha-yr</b>	10%	15%	20%	25%	30%		
2	100%	100%	100%	100%	100%		
3	100%	100%	100%	100%	100%		
4	95%	100%	100%	100%	100%		
5	92%	98%	100%	100%	100%		
6	88%	98%	100%	100%	100%		
7	87%	97%	100%	100%	100%		
8	82%	91%	99%	100%	100%		
9	81%	91%	99%	100%	100%		
10	78%	89%	97%	99%	99%		
11	76%	88%	96%	99%	99%		
12	73%	86%	94%	96%	96%		
13	73%	85%	94%	96%	96%		
14	70%	83%	92%	94%	95%		
15	69%	81%	90%	92%	93%		
16	68%	81%	89%	91%	92%		
17	68%	81%	89%	91%	92%		
18	67%	79%	87%	89%	90%		



2	Figure 5A-45.	Cumulative percent of ecoregions with exceedances >10, >15, >20, >25,
3	-	>30% as a function of total S deposition across all 5 deposition periods
4		(2001-03, 2006-08, 2010-12, 2014-06, 2018-20). 100% indicates there were
5		no ecoregion that had percent exceedances above >10, >15, >20, >25, >30%
6		for a given deposition level. Critical load exceedances based on ANC
7		threshold of 30 µeq/L for the eastern U.S. (See Table 5A-28 for data).
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### 1Table 5A-30.Number of ecoregions with percent of exceedances of >10, >15, >20, >25,2>30% as a function of total S deposition across all 5 deposition periods3(2001-03, 2006-08, 2010-12, 2014-06, 2018-20). Includes 18 ecoregions in4the eastern U.S. and Critical load exceedances are based on an ANC5threshold of 20 μeq/L.

Total Sulfur Deposition	Number of Ecoregion with Percent of Exceedances Across the 5 deposition Periods							
(Kg S/ha-yr)	10%	15%	20%	25%	30%			
2	0	0	0	0	0			
3	0	0	0	0	0			
4	0	0	0	0	0			
5	2	1	0	0	0			
6	4	1	0	0	0			
7	5	1	0	0	0			
8	9	4	0	0	0			
9	9	4	0	0	0			
10	11	6	1	1	1			
11	13	7	2	1	1			
12	15	9	4	3	2			
13	16	10	4	3	2			
14	19	12	6	4	3			
15	21	14	8	6	4			
16	22	15	9	7	5			
17	22	15	9	7	5			
18	24	17	11	9	7			
19	24	17	11	9	7			
20	24	17	11	9	7			

# 1Table 5A-31.Cumulative percent of waterbodies in ecoregions meeting the target ANC2values as a function of total S deposition across all 5 deposition periods3(2001-03, 2006-08, 2010-12, 2014-06, 2018-20).100% indicates there were4no ecoregions that had percent exceedances above >10, >15, >20, >25,5>30% for a given deposition level. Critical load exceedances based on ANC6threshold of 20 μeq/L for the eastern U.S. (See Table 5A-29 for data).

Total Sulfur Deposition	Number of the 5 depo	f Ecoregion v sition Period	vith Percent o Is	of Exceedanc	es Across
(Kg S/ha-yr)	10%	15%	20%	25%	30%
2	100%	100%	100%	100%	100%
3	100%	100%	100%	100%	100%
4	100%	100%	100%	100%	100%
5	96%	98%	100%	100%	100%
6	93%	98%	100%	100%	100%
7	92%	98%	100%	100%	100%
8	87%	94%	100%	100%	100%
9	87%	94%	100%	100%	100%
10	85%	92%	99%	99%	99%
11	83%	91%	97%	99%	99%
12	81%	89%	95%	96%	97%
13	80%	88%	95%	96%	98%
14	77%	86%	93%	95%	96%
15	76%	84%	91%	93%	95%
16	75%	83%	90%	92%	94%
17	75%	83%	90%	92%	94%
18	73%	81%	88%	90%	92%



Figure 5A-46.	Cumulative percent of ecoregions with exceedances >10, >15, >20, >25,					
	>30% as a function of total S deposition across all 5 deposition periods					
	(2001-03, 2006-08, 2010-12, 2014-06, 2018-20). 100% indicates there were					
	no ecoregion that had percent exceedances above >10, >15, >20, >25, >30%					
	for a given deposition level. Critical load exceedances based on ANC					
	threshold of 20 µeq/L for the eastern U.S. (See Table 5A-30 for data).					

1Table 5A-32.Number of ecoregions with percent of exceedances of >10, >15, >20, >25,2>30% as a function of total S deposition across all 5 deposition periods3(2001-03, 2006-08, 2010-12, 2014-06, 2018-20). Includes 7 ecoregions in the4western U.S. and critical load exceedances are based on ANC threshold of520 μeq/L.

Total Sulfur	Number of Ecoregion with Percent of Exceedances Across the 5 deposition Periods					
Kg S/ha-yr	10%	15%	20%	25%	30%	
2	0	0	0	0	0	
3	0	0	0	0	0	
4	0	0	0	0	0	
5	0	0	0	0	0	
6	0	0	0	0	0	
7	0	0	0	0	0	
8	0	0	0	0	0	
9	0	0	0	0	0	
10	0	0	0	0	0	
11	0	0	0	0	0	
12	0	0	0	0	0	
13	0	0	0	0	0	
14	0	0	0	0	0	
15	0	0	0	0	0	
16	0	0	0	0	0	
17	0	0	0	0	0	
18	0	0	0	0	0	
19	0	0	0	0	0	

1 Table 5A-33. Cumulative percent of waterbodies in ecoregions meeting the target ANC values as a function of total S deposition across all 5 deposition periods (2001-03, 2006-08, 2010-12, 2014-06, 2018-20). 100% indicates there were no ecoregions that had percent exceedances above >10, >15, >20, >25, >30% for a given deposition level. Critical load exceedances based on ANC threshold of 20 µeq/L for the western U.S. (See Table 5A-31 for data).

Total Sulfur Deposition	Number	ber of Ecoregion with Percent of Exceedances Across the 5 deposition Periods					
Kg S/ha-yr	10%	15%	20%	25%	30%		
2	100%	100%	100%	100%	100%		
3	100%	100%	100%	100%	100%		
4	100%	100%	100%	100%	100%		
5	100%	100%	100%	100%	100%		
6	100%	100%	100%	100%	100%		
7	100%	100%	100%	100%	100%		
8	100%	100%	100%	100%	100%		
9	100%	100%	100%	100%	100%		
10	100%	100%	100%	100%	100%		
11	100%	100%	100%	100%	100%		
12	100%	100%	100%	100%	100%		
13	100%	100%	100%	100%	100%		
14	100%	100%	100%	100%	100%		
15	100%	100%	100%	100%	100%		
16	100%	100%	100%	100%	100%		
17	100%	100%	100%	100%	100%		
18	100%	100%	100%	100%	100%		
19	100%	100%	100%	100%	100%		



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threshold of 20 µeq/L for the western U.S. (See Table 5A-33 for data).

# 1Table 5A-34.Number of ecoregions with percent of exceedances of >10, >15, >20, >25,2>30% as a function of total S deposition across all 5 deposition periods3(2001-03, 2006-08, 2010-12, 2014-06, 2018-20). Includes 25 ecoregions4across the U.S. and critical load exceedances are based on ANC threshold5of 50 µeq/L for the east and 20 µeq/L for the west.

Total Sulfur	Number of Ecoregion with Percent of Exceedances Across the 5 deposition Periods				
Kg S/ha-yr	10%	15%	20%	25%	30%
2	0	0	0	0	0
3	1	0	0	0	0
4	3	1	0	0	0
5	9	3	2	1	0
6	13	4	2	1	0
7	14	5	3	1	0
8	18	9	5	3	0
9	19	9	5	3	0
10	22	11	6	4	1
11	24	13	7	4	1
12	27	15	9	6	3
13	28	16	10	6	3
14	31	18	12	8	5
15	33	20	14	10	7
16	34	21	15	11	8
17	34	21	15	11	8
18	36	23	17	13	10
19	36	23	17	13	10

1 Table 5A-35. Cumulative percent of waterbodies in ecoregions meeting the target ANC values as a function of total S deposition across all 5 deposition periods (2001-03, 2006-08, 2010-12, 2014-06, 2018-20). 100% indicates there were no ecoregions that had percent exceedances above >10, >15, >20, >25, >30% for a given deposition level. Critical load exceedances based on ANC threshold of 50 µeq/L for the east and 20 µeq/L for the west (See Table 5A-33 for data).

Total Sulfur Deposition	Number of Ecoregion with Percent of Exceedances Across the 5 deposition Periods						
Kg S/ha-yr	10%	15%	20%	25%	30%		
2	100%	100%	100%	100%	100%		
3	98%	100%	100%	100%	100%		
4	96%	99%	100%	100%	100%		
5	90%	97%	98%	99%	100%		
6	86%	96%	98%	99%	100%		
7	86%	95%	97%	99%	100%		
8	82%	91%	95%	97%	100%		
9	82%	91%	95%	97%	100%		
10	80%	90%	94%	96%	99%		
11	78%	88%	94%	96%	99%		
12	76%	87%	92%	95%	97%		
13	76%	86%	91%	95%	97%		
14	74%	85%	90%	93%	96%		
15	73%	83%	88%	92%	94%		
16	72%	83%	88%	91%	93%		
17	72%	83%	88%	91%	93%		
18	71%	82%	86%	90%	92%		



2	Figure 5A-48.	Cumulative percent of ecoregions with exceedances >10, >15, >20, >25,
3		>30% as a function of total S deposition across all 5 deposition periods
4		(2001-03, 2006-08, 2010-12, 2014-06, 2018-20). 100% indicates there were
5		no ecoregion that had percent exceedances above >10, >15, >20, >25, >30%
6		for a given deposition level. Critical load exceedances based on ANC
7		threshold of 50 µeq/L for the east and 20 µeq/L for the west (50/20 µeq/L)
8		(See Table 5A-35 for data).

# 1Table 5A-36.Number of ecoregions with percent of exceedances of >10, >15, >20, >25,2>30% as a function of total S deposition across all 5 deposition periods3(2001-03, 2006-08, 2010-12, 2014-06, 2018-20). Includes 25 ecoregions4across the U.S. and critical load exceedances are based on ANC threshold5of 30 µeq/L for the east and 20 µeq/L for the west.

Total Sulfur Deposition	Number of Ecoregion with Percent of Exceedances Across the 5 deposition Periods				
Kg S/ha-yr	10%	15%	20%	25%	30%
2	0	0	0	0	0
3	0	0	0	0	0
4	2	0	0	0	0
5	4	1	0	0	0
6	7	1	0	0	0
7	8	2	0	0	0
8	12	6	1	0	0
9	13	6	1	0	0
10	16	8	2	1	1
11	18	9	3	1	1
12	21	11	5	3	3
13	22	12	5	3	3
14	25	14	7	5	4
15	27	16	9	7	6
16	28	17	10	8	7
17	28	17	10	8	7
18	30	19	12	10	9
19	30	19	12	10	9

1Table 5A-37.Cumulative percent of waterbodies in ecoregions meeting the target ANC values2as a function of total S deposition across all 5 deposition periods (2001-03, 2006-308, 2010-12, 2014-06, 2018-20).100% indicates there were no ecoregions that4had percent exceedances above >10, >15, >20, >25, >30% for a given5deposition level. Critical load exceedances based on ANC threshold of 306µeq/L for the east and 20 µeq/L for the west (See Table 5A-35 for data).

Total Sulfur Deposition	Numbe	r of Ecoregion with Percent of Exceedances Across the 5 deposition Periods				
Kg S/na-yr	10%	15%	20%	25%	30%	
2	100%	100%	100%	100%	100%	
3	100%	100%	100%	100%	100%	
4	97%	100%	100%	100%	100%	
5	95%	99%	100%	100%	100%	
6	93%	99%	100%	100%	100%	
7	92%	98%	100%	100%	100%	
8	88%	94%	99%	100%	100%	
9	88%	94%	99%	100%	100%	
10	85%	93%	98%	99%	99%	
11	84%	92%	97%	99%	99%	
12	82%	90%	96%	97%	97%	
13	81%	90%	96%	97%	97%	
14	79%	88%	94%	96%	97%	
15	78%	87%	93%	94%	95%	
16	77%	86%	92%	93%	94%	
17	77%	86%	92%	93%	94%	
18	76%	85%	90%	92%	93%	



2	Figure 5A-49.	Cumulative percent of ecoregions with exceedances >10, >15, >20, >25,
3		>30% as a function of total S deposition across all 5 deposition periods
4		(2001-03, 2006-08, 2010-12, 2014-06, 2018-20). 100% indicates there were
5		no ecoregion that had percent exceedances above >10, >15, >20, >25, >30%
6		for a given deposition level. Critical load exceedances based on ANC
7		threshold of 30 µeq/L for the east and 20 µeq/L for the west (50/20 µeq/L)
8		(See Table 5A-37 for data).

## 1Table 5A-38.Number of ecoregions with percent of exceedances of >10, >15, >20, >25,2>30% as a function of total S deposition across all 5 deposition periods3(2001-03, 2006-08, 2010-12, 2014-06, 2018-20). Includes 25 ecoregions4across the U.S. and critical load exceedances are based on ANC threshold50f 20 µeq/L for both the east the west U.S.

Total Sulfur Deposition	Number of Ecoregion with Percent of Exceedances Across the 5 deposition Periods				
Kg S/ha-yr	10%	15%	20%	25%	30%
2	0	0	0	0	0
3	0	0	0	0	0
4	0	0	0	0	0
5	2	1	0	0	0
6	4	1	0	0	0
7	5	1	0	0	0
8	9	4	0	0	0
9	9	4	0	0	0
10	11	6	1	1	1
11	13	7	2	1	1
12	15	9	4	3	2
13	16	10	4	3	2
14	19	12	6	4	3
15	21	14	8	6	4
16	22	15	9	7	5
17	22	15	9	7	5
18	24	17	11	9	7
19	24	17	11	9	7
1Table 5A-39.Cumulative percent of waterbodies in ecoregions meeting the target ANC values2as a function of total S deposition across all 5 deposition periods (2001-03, 2006-308, 2010-12, 2014-06, 2018-20). 100% indicates there were no ecoregions that4had percent exceedances above >10, >15, >20, >25, >30% for a given5deposition level. Critical load exceedances based on ANC threshold of 206µeq/L for the east and west U.S. (See Table 5A-37 for data).

Total Sulfur Deposition	Number of Ecoregion with Percent of Exceedances Across the 5 deposition Periods							
Kg S/ha-yr	10%	15%	20%	25%	30%			
2	100%	100%	100%	100%	100%			
3	100%	100%	100%	100%	100%			
4	100%	100%	100%	100%	100%			
5	98%	99%	100%	100%	100%			
6	96%	99%	100%	100%	100%			
7	95%	99%	100%	100%	100%			
8	91%	96%	100%	100%	100%			
9	91%	96%	100%	100%	100%			
10	90%	94%	99%	99%	99%			
11	88%	94%	98%	99%	99%			
12	87%	92%	96%	97%	98%			
13	86%	91%	97%	97%	98%			
14	84%	90%	95%	97%	97%			
15	83%	88%	93%	95%	97%			
16	82%	88%	93%	94%	96%			
17	82%	88%	93%	94%	96%			
18	81%	86%	91%	93%	94%			



2	Figure 5A-50.	Cumulative percent of ecoregions with exceedances >10, >15, >20, >25,
3	_	>30% as a function of total S deposition across all 5 deposition periods
4		(2001-03, 2006-08, 2010-12, 2014-06, 2018-20). 100% indicates there were
5		no ecoregion that had percent exceedances above >10, >15, >20, >25, >30%
6		for a given deposition level. Critical load exceedances based on ANC
7		threshold of 20 $\mu$ eq/L for the east and west U.S. (See Table 5A-39 for data).



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### 5A.2.3 Analysis of Risk in Case Study Areas for Acidification

The case study areas represent geographic diverse acid sensitive areas across the CONUS that have sufficient data to complete a quantitative analysis. This includes the necessary air quality information to assess varying levels of deposition, including monitoring and deposition information. In addition, the deposition levels across these set of case studies should generally reflect the range of concentration and deposition levels found across the CONUS. Five case study areas were identified that meet the criteria (Figure 5A-52), 3 in the eastern U.S. (NOMN,

8 SHVA and WHMT) and 2 areas are in the western U.S. (GILA, ROMO and SINE).



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Figure 5A-52. Location of the case study areas. Northern Minnesota (NOMN), Rocky
 Mountain National Park (ROMO), Shenandoah Valley (SHVA), Sierra
 Nevada Mountains (SINE) and White Mountain National Forest (WHMT).

13 This section presents a summary of the CL assessment for deposition scenarios

- 14 representing just meeting the current annual secondary PM<sub>2.5</sub> NAAQS. Using the same
- 15 methodology described in section 5A.1.2 above, this assessment estimated potential CL
- 16 exceedances for three air quality (AQ) scenarios. Not all case study areas had sufficient water

- quality or aquatic CL data to provide an in-depth analysis. Aquatic CLs for the 5 case study areas
   are summarized below using the following steps:
- 3 (1) CLs were extracted from the NCLD for each of the 5 case study areas for the following
   4 ANC thresholds: 20, 30, 50, and 80 μeq/L.
- (2) CLs were summarized for each area in terms of the average, 70th and 90th percentile.
   This was done in terms of kg S/ha-yr for S only analyses and meq/m<sup>2</sup>-yr for N and S analyses.
- 8 (3) Exceedances were calculated for each of the AQ scenarios for all 4 ANC thresholds for S
   9 only and N+S.
- (4) The exceedances were summarized as the percent of waterbodies that were exceeding in
   each area for all CLs and for the 70th and 90th percentiles.
- 12 **5A.2.3.1 Results**

13 A total of 524 CLs were found in the 5 case study areas, excluding SHVA which had 14 complete coverage (4977 Total CL with 704 sensitive CLs). ROMO, SINE, NOMN, and 15 WHMT had 121, 139, 183, and 74 CLs respectively (Figure 5A-53). Despite the relatively high 16 number of aquatic CLs for these four case studies, they do not represent a complete coverage of 17 water resources and the summary of the CLs and exceedances only represent the waterbodies 18 that have been modelled. Table 5A-40 provides average, 70th and 90th percentile CLs for S only 19 for each case study areas in units of Kg S/ha-yr. Table 5A-41 also provides the same 20 information but also includes CLs for S and N but in units of meq/m<sup>2</sup>-yr. Critical loads for S only 21 were found to be similar for the waterbodies modelled among the case study areas with higher 22 CL values for the lower ANC thresholds. The below summary is based on an ANC threshold of 23  $50 \mu \text{eq/L}$ . Average S only CL values range from 6.6 to 9.8 kg S/ha-yr or 41.3 to 61.3 meq/m<sup>2</sup>-yr 24 for waterbodies with CLs within each of the case study areas. The 90th percentile CLs for S 25 only are similar among the case studies and range between 0.1 to 4.1 kg S/ha-yr or 0.1 to 25.8 26  $meq/m^2$ -yr.

Table 5A-40. Average, 70<sup>th</sup> and 90<sup>th</sup> percentile CL of S only (kg S/ha-yr) for each case
 study area for ANC limits of 20, 30, 50, and 80 µeq/L.

	20 µeq/L			30 µeq/L			50 µeq/L			80 µeq/L		
	Ave.	70th	90th									
Sulfur (S) only												
ROMO	9.5	5.4	3.6	8.5	4.5	2.6	6.6	2.7	0.5	4.3	0.1	0.1
SINE	12.0	4.1	1.8	11.0	2.8	0.5	9.3	0.6	0.1	7.5	0.1	0.1
NOMN	10.8	5.5	4.2	10.4	5.3	3.9	9.8	4.7	3.2	8.2	3.8	2.3
WHMT	10.6	6.9	4.4	9.6	6.1	3.3	7.4	4.1	0.7	4.7	0.4	0.1
SHVA	12.4	9.4	7.1	11.4	8.4	6.3	9.4	6.3	4.1	6.6	3.2	1.3

	20 µeq/L				30 µeq/L 50 µec			50 µeq/L	L 80 µeq/L			
	Ave.	70th	90th	Ave.	70th	90th	Ave.	70th	90th	Ave.	70th	90th
	Sulfur (S) only											
ROMO	59.1	34.0	22.6	5.30	28.4	16.1	41.2	16.7	3.4	26.7	0.1	0.1
SINE	75.0	25.4	11.0	68.7	17.3	2.9	58.4	3.5	0.1	47.1	0.1	0.1
NOMN	67.4	34.5	26.0	65.3	32.4	29.1	61.0	29.3	20.1	54.6	23.8	14.4
WHMT	66.3	43.4	27.8	59.7	38.3	20.8	46.3	25.6	4.4	29.6	2.3	0.1
SHVA	77.4	58.9	44.6	71.3	52.4	39.1	59	39.5	25.8	41.4	20.3	8.0
				Sulfu	ur and N	litrogen (	N and S)					
ROMO	100.9	76.2	63.1	94.8	70.1	57.3	83.1	58.8	46.6	68.5	46.1	37.9
SINE	120.4	66.9	49.6	114.1	61.1	42.6	103.8	47.8	38.8	92.4	41.7	28.0
NOMN	110.4	76.0	67.7	108.3	74.2	65.9	104.0	70.4	62.1	97.6	64.7	56.2
WHMT	104.4	83.7	68.5	97.6	75.1	59.7	84.1	62.1	47.9	81.2	47.0	37.6
SHVA												

1Table 5A-41.Average, 70th and 90th percentile CL of S and S+N (meq/m²-yr) for each2case study area for ANC limits of 20, 30, 50, and 80 μeq/L.



### Figure 5A-53. Critical load maps of each case study area. Critical load for sulfur (S) using an ANC threshold is mapped with units of meq/m<sup>2</sup>-yr. Upper left to right is Rocky Mountain National Park (ROMO), Northern Minnesota (NOMN), and White Mountains National Forest (WHMT). Lower left to right is Sierra Nevada Mountains, (SINE) and Shenandoah Valley Area (SHVA).

7 Critical load exceedances were calculated for several air quality scenarios that reflected 8 an area meeting the most controlling<sup>2</sup> current secondary NAAQS for that area (of those for SO<sub>2</sub>, 9 NO<sub>2</sub> and PM), which in all cases was that for PM<sub>2.5</sub>. For each case study area, historic air quality 10 was examined to find a time when the monitors within or near the area influencing the case study 11 area<sup>3</sup> had design values that were within 10% of the current standard level (i.e. 15  $\mu$ g/m<sup>3</sup>). To

12 examine how changing air quality and corresponding deposition could affect these estimated

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 $<sup>^2</sup>$  The scenarios selected had air quality for which the PM<sub>2.5</sub> design value for the highest monitor was just equal to the current secondary standard.

<sup>&</sup>lt;sup>3</sup> The area of influence is defined as the region where a change in emissions leads to a change in deposition at the case study area. A recent study of Class I areas found that the area of influence for nitrogen deposition can vary, and the radius was estimated to range between 500 – 1200 km (Lee et al., 2016). To ensure that emissions and concentrations in the area of influence are relevant, this analysis uses a maximum radius of 500 km.

- 1 exceedances, additional scenarios for air quality at these locations in other years were also
- $2 \qquad \text{analyzed with the aim of having similar maximum PM_{2.5} annual design values across the case}$
- 3 studies. For these additional scenarios, time periods were selected where the highest monitor in
- 4 the area of influence was within 10% of 12  $\mu$ g/m<sup>3</sup> and 10  $\mu$ g/m<sup>3</sup>. For each of the selected air
- 5 quality periods, the TDEP data were extracted for S and N. For one case study area, SINE,<sup>4</sup> the
- 6 air quality and TDEP data were adjusted slightly downwards to reflect a relevant air quality

7 scenario. For some locations, it was not possible to select a three-year historical period as PM<sub>2.5</sub>

8 concentrations, currently and in the past, have not been as high as the threshold for that scenario.

9 The air quality periods analyzed, and associated deposition levels are shown in Table 5A-42 and

10 Table 5A-43.

Case Study Area	TDEP years for 15 µg m-3	TDEP years for 12 µg m-3	TDEP years for 10 µg m-3
Coastal South Carolina	2004-2006	2007-2009	2011-2013
Gila National Forest	PM2.5 concentrations have not been this high	2002-2004	2005-2007
Northern Minnesota	PM2.5 concentrations have not been this high	2000—2002	2007-2009
Rocky Mountain National Park	PM2.5 concentrations have not been this high	PM2.5 concentrations have not been this high	2000-2002
Shenandoah Valley	2005—2007	2009—2011	2014-2016
Sierra Nevada	S deposition: 0.70 N deposition: 0.72	S deposition: 0.56 N deposition: 0.57	S deposition: 0.46 N deposition: 0.48
White Mountain National Forest	2000—2002	2005-2007	2009-2011

11 Table 5A-42. The three-year historical periods used for each case study area.

<sup>&</sup>lt;sup>4</sup> For the Sierra Nevada case study, there is no historical period that is at or near the target PM<sub>2.5</sub> concentration, so it is not possible to use a historical dataset of deposition. Instead, this assessment approximates the change in deposition due to a change in PM<sub>2.5</sub> concentration at the maximum monitor. A linear model was fit using air concentration and total (wet + dry) deposition from a 21-year CMAQ model simulation. First, the air concentration and deposition values were normalized by their mean value. A linear model was fit to predict total deposition from air concentration. The slope was an estimate of the change in deposition due to a change in PM<sub>2.5</sub> concentration. The slope was an estimate of the change in deposition when the PM<sub>2.5</sub> concentration at the highest monitor was reduced to 10 μg m<sup>-3</sup>, 12 μg m<sup>-3</sup>, and 15 μg m<sup>-3</sup>. The prediction interval at each of these concentration levels was 40%, which indicates that there are a range of deposition levels that are consistent with these air concentration targets. The predicted deposition change for nitrogen and sulfur were different by a small amount, which reflects differences in the relationship between PM<sub>2.5</sub> and deposition. To be clear, this is not meant to be a prediction, but rather a plausible deposition scenario associated with maximum PM<sub>2.5</sub> concentrations for each target level.

#### 1 **Table 5A-43.** 1 2 **a** 3 **b**

For each three-year period described in Table 5A-41, this is the three-year average deposition, spatially averaged across the case study area, for N and S deposition. These values are calculated from the TDEP dataset.

	Mean N dep	oosition (kg N	I ha⁻¹ year⁻¹)	Mean S deposition (kg S ha <sup>-1</sup> year <sup>-1</sup> )			
Case study	15 µg/m³	12 µg/m³	10 µg/m³	15 µg/m³	12 µg/m³	10 µg/m³	
Northern Minnesota	NA	6.8	6.0	NA	3.4	3.0	
Rocky Mountain National Park	NA	NA	6.6	NA	NA	2.3	
Shenandoah National Park	11	8.7	8.3	10	5.0	3.1	
Sierra Nevada	4.9*	3.9*	3.3*	0.80*	0.64*	0.53*	
White Mountain National Forest (New Hampshire)	7.6	6.7	5.2	7.2	7.1	3.8	

\*The air quality and associated deposition estimates for Sierra Nevada case study are based on a "roll down" approach. The highest PM<sub>2.5</sub> DVs in the area were rolled down to equal the specified value for each scenario (15, 12 and 10  $\mu$ g/m<sup>3</sup>) and a unit S or N deposition per unit PM<sub>2.5</sub> concentration (from a regression based on 21-year CMAQ simulation) was applied to derive the associated deposition estimates presented here.

4 The case study areas, when taken as a group, represent a large range of PM<sub>2.5</sub> and 5 deposition conditions. For all case study areas, the correlation between the sulfate PM<sub>2.5</sub> and 6 nitrate PM<sub>2.5</sub> measured within the case study area and the PM<sub>2.5</sub> monitor that measures the 7 highest concentrations within the area of influence are shown in Table 5A-44. The measured 8 sulfate and nitrate PM<sub>2.5</sub> is found to be highly correlated with the maximum PM<sub>2.5</sub> monitor values 9 for these case study areas. Table 5A-44 also lists the correlation between wet deposition and air 10 concentrations within the case study area. Wet deposition of sulfur is correlated with air 11 concentrations of sulfur in the eastern U.S. case study areas, but the correlation is absent for 12 some case study areas in the West. Correlations for air concentration of nitrate PM<sub>2.5</sub> and wet 13 deposition of nitrogen are low for most case study areas. The case study areas in the western U.S. 14 have greater inter-annual variability in precipitation, which adds variability to the air 15 concentration-deposition relationship. Furthermore, the measured wet deposition provides only 16 part of the deposition budget; dry deposition is not routinely measured, and models are needed to 17 complete the assessment of the air concentration-deposition relationship. 18 The relationship between air concentration and deposition depends on several factors, 19 including the chemical form of sulfur and nitrogen, the vertical distribution in the atmosphere, 20 and the frequency of precipitation. Each of these vary across the different case study areas. For 21 case study areas in the eastern U.S., where SO<sub>2</sub> and NOx emissions have declined the most, 22 measurements of PM2.5 within the area of influence and wet deposition within the case study area 23 show a strong correlation. For case study areas in the western U.S., where dry deposition and 24 ammonia play a larger role, in some cases there is no correlation between measured wet

25 deposition and surface concentrations.

1 As some locations do not show a correlation of air concentrations and wet deposition, it is 2 necessary to investigate further with models which can also estimate the dry deposition 3 component. Table 5A-45 shows the correlation calculated between simulated PM<sub>2.5</sub> 4 concentrations at each case study area, where the annual average concentration and annual total 5 deposition are shown as calculated by a 21-year CMAQ simulation (Zhang et al., 2018). The 6 CMAQ simulation provides a more complete quantification of the relationship between air 7 concentrations and deposition because both wet and dry deposition are included. However, the 8 model inherently lacks some of the variability that arises from making measurements of air 9 concentration in the field. Nevertheless, the correlations between the CMAQ-simulated annual 10 average air concentration and total annual deposition are higher than the comparisons between 11 observed air concentration and wet deposition, which suggests the air concentrations and 12 deposition are more tightly linked than can be estimated from the observational dataset, which 13 does not include dry deposition.

14Table 5A-44.Summary of correlation between observations of air concentration and15NADP deposition.

Case Study Areas	Correlation between sulfate PM and total PM <sub>2.5</sub> mass	Correlation between nitrate PM and total PM <sub>2.5</sub> mass	Correlation between wet deposition and total sulfur air concentrations	Correlation between wet deposition and total nitrate air concentrations
Northern Minnesota	0.95	0.87	0.78	0.07
Rocky Mountain National Park	0.86	0.92	0.41	0.13
Shenandoah Valley Area	0.99	0.96	0.93	0.71
Sierra Nevada Mountains	0.78	0.85	-0.03	-0.08
White Mountain National Forest	0.99	0.88	0.81	0.61

1Table 5A-45.Correlation between CMAQ-simulated annual sum of total deposition and<br/>the CMAQ-simulated annual average concentration for each case study.3The correlation is calculated by computing the annual average<br/>concentration and annual total deposition from 21-year CMAQ simulation<br/>(Zhang et al., 2018). This table compares total (wet + dry) deposition and<br/>air concentrations.

Correlation between:						
Case Study Area	Total nitrate air concentration and N deposition	Total sulfur air concentration and S deposition				
Northern Minnesota	0.42	0.71				
Rocky Mountain National Park	0.60	0.68				
Shenandoah Valley	0.93	0.88				
Sierra Nevada: Sequoia National Park	0.94	0.74				
White Mountain National Forest	0.80	0.91				

7 For the N and S deposition associated with these air quality scenarios, critical load

8 exceedances were calculated for S, and for N and S combined, for each waterbody in each case

9 study area. Exceedances for N and/or S were calculated for all case study areas except for

10 SHVA. Table 5A-46 contains percent exceedances (number waterbodies exceeding the CL

11 divided by the total number of waterbodies with CLs in the case study area times 100) and the

12 absolute number of waterbodies that exceed the CL. All four ANC thresholds were evaluated.

13 Unlike the CLs, exceedances are not consistent among the case study areas. Percent exceedances

14 were similar between CL values determined for S only and for N and/or S deposition. The

15 highest percent exceedances occurred for the ANC limit of 80 µeq/L while lower percent

16 exceedances occurred for ANC of 20 µeq/L, as expected, for all scenarios.

Air Quality		Sulf	ur Only	Sul Ni	fur and trogen	Sulf	ur Only	Sulf Nitr	ur and rogen	Sulf	ur Only	Sulf Nit	ur and rogen	Sulf	ur Only	Sulf Niti	ur and rogen
Scenario	Areas	No.	Percent	No.	Percent	No.	Percent	No.	Percent	No.	Percent	No.	Percent	No.	Percent	No.	Percent
µg/m₃		A	NC Target	= 20 µ	eq/L	A	NC Target	t = 30 µ	ieq/L	A	NC Targe	t = 50 µ	eq/L	Α	NC Targe	t = 80 µ	ieq/L
	ROMO	3	2%	6	5%	6	5%	16	13%	25	21%	37	31%	60	50%	69	57%
	SINE	1	1%	1	1%	3	2%	3	2%	13	9%	13	9%	33	24%	33	24%
10	NOMN	2	1%	2	1%	2	1%	2	1%	3	2%	4	2%	16	9%	16	9%
	WHMT	3	4%	5	7%	9	12%	10	14%	18	24%	19	26%	36	49%	39	53%
	SHVA	9	2%			11	2%			20	4%			107	23%		
	ROMO																
	SINE	1	1%	1	1%	9	6%	9	6%	34	24%	34	24%	61	44%	61	44%
12	NOMN	2	1%	6	3%	2	1%	11	6%	6	3%	21	11%	27	15%	47	26%
	WHMT	21	28%	30	41%	25	33%	36	49%	37	50%	48	65%	48	65%	57	77%
	SHVA	16	3%			19	4%			68	15%			192	41%		
	ROMO																
	SINE	2	1%	2	1%	11	8%	11	8%	38	27%	38	27%	62	45%	62	45%
15	NOMN																
	WHMT	23	31%	35	47%	27	36%	41	55%	38	51%	49	66%	48	64%	61	82%
	SHVA	156	34%			202	44%			279	60%			366	79%		

1	Table 5A-46.	Number and percent of case study waterbodies estimated to exceed their CLs for specified ANC targets and
2		air quality scenario.

The results are summarized in Table 5A-47 for each of the case study areas. Across the
 case studies and ANC thresholds, S deposition would need to be on average between 7.4 to 12
 kg/ha-yr and 4.1 to 9.4 and 0.7 to 7.1 to attain a 70 and 90 percentile, respectively.

		Eastern		Wes	stern					
ANC (µeq/L)	Northern MN	White Mtns	Shenandoah	Rocky Mtns	Sierra NV Mtns					
	Based on Averaging of All Sites Achieving 20 µeq/L									
20	11	11	12	9.5	12					
30	10	10	11							
50	10	7.4	9.4							
	Based on 70% of sites Achieving 30 µeq/L									
20	5.5	6.9	9.4	5.4	4.1					
30	5.3	6.1	8.4							
50	4.7	4.1	6.3							
		Based o	n 90% of sites Ach	nieving 50 µeq/L						
20	4.2	4.4	7.1	3.6	1.8					
30	3.9	3.3	6.3							
50	3.2	0.7	4.1							

### Table 5A-47. Summary of S deposition levels to attain an ANC target of 20, 30, and 50 μeq/L for case study areas.

### 6 5A.3 KEY UNCERTAINTIES/LIMITATIONS

7 There is uncertainty associated with the parameters in the steady-state critical load model 8 used to estimate aquatic CLs. The strength of the CL estimate and the exceedance calculation 9 relies on the ability to estimate the catchment-average base-cation supply (i.e., input of base 10 cations from weathering of bedrock and soils and air), runoff, and surface water chemistry. The 11 uncertainty associated with runoff and surface water measurements is well known. However, the 12 ability to accurately estimate the catchment supply of base cations to a waterbody can be 13 uncertain. This is important because the catchment supply of base cations from the weathering of 14 bedrock and soils is the factor that has the most influence on the CL calculation and also has the 15 largest uncertainty (Li and McNulty, 2007). Although the approach to estimate base-cation 16 supply for the national case study (e.g., F-factor approach) has been widely published and 17 analyzed in Canada and Europe, and has been applied in the CONUS (e.g., Dupont et al., 2005 18 and others), the uncertainty in this estimate is unclear and could be large in some cases. For this 19 reason, an uncertainty analysis of the state-steady CL model was completed to evaluate the 20 uncertainty in the CL and exceedance estimation. 21 A probabilistic analysis using a range of parameter uncertainties was used for CLs

determined by the SSWC model using the F-factor approach to assess (1) the confidence interval

- of the CL, (2) the degree of confidence in the exceedance values and (3) coefficient of variation
- 24 (CV) of the critical load. The probabilistic framework is Monte Carlo, whereby each steady-state

- 1 input parameter varies according to specified probability distributions and their range of
- 2 uncertainty (Table 5A-48). The purpose of the Monte Carlo methods was to propagate the
- 3 uncertainty in the model parameters in the steady-state CL model.

# Table 5A-48. Parameters used and their uncertainty range. The range of surface water parameters (e.g., CA, MG, CL, NA, NO<sub>3</sub>, SO<sub>4</sub>) were determined from surface water chemistry. Runoff(Q) based on min. and max value from long-term water quality data. Acidic Deposition were set at 25%.

Parameter	Units	Uncertainty range	Distribution
Q	m/yr	1971-2000 annual runoff	Normal
CA	µeq/L	Min. and Max	Normal
MG	µeq/L	Min. and Max	Normal
CL	µeq/L	Min. and Max	Normal
NA	µeq/L	Min. and Max	Normal
NO <sub>3</sub>	µeq/L	Min. and Max	Normal
SO <sub>4</sub>	µeq/L	Min. and Max	Normal
Acidic Deposition (NOx & SO <sub>4</sub> )	meq/m <sup>2</sup> -yr	25%	Lognormal

8 Within the Monte Carlo analysis, model calculations were run enough times (i.e. 5,000

9 times) to capture the range of behaviors represented by all SSWC model parameters (Table 5A-

10 48). The parameter uncertainty ranges were determined by various methods. For runoff (Q), the

11 1971-2016 annual runoff (m/yr) (Wieczorek et al. 2018) was used for each waterbody. Water

12 quality uncertainty range was based on the minimum and maximum data range for a waterbody

13 where 6-years of water quality data exits. For waterbodies with insufficient water quality data,

14 the minimum and maximum range was based on a range determined from regional long-term

15 water quality data from the EPA's Long-term Monitoring (LTM) program

16 (<u>https://www.epa.gov/airmarkets/monitoring-surface-water-chemistry</u>) and other local programs.

17 Regions were defined for New England, Adirondacks, Central Appalachia Mountains, and Mid-

18 Alantic. The Monte Carlo analysis was done in R. A total of 14,943 waterbodies in the CONUS

19 were analyzed (Figure 5A-54).

20 The magnitude of the error for the N leaching (method A) was determined by quantifying

21 the uncertainty of the flux of nitrate (NO<sub>3</sub><sup>-</sup>) to a given lake or stream. Water quality data for the

22 past 28 years from the EPA's Long-term Monitoring (LTM) program was used to assess the

23 uncertainty of the influx of nitrate (NO<sub>3</sub><sup>-</sup>). Lakes or streams are sampled weekly to quarterly

24 depending on the site and program. Annual flux of nitrate was calculated using annual

- 25 concentration of  $NO_3^-$  for a given monitoring site and multiplied by annual runoff (m/yr)
- 26 (Wieczorek et al. 2018) for the watershed and year. Confidence intervals were calculated for

- 1 monitoring sites for a given region (i.e., New England, Adirondacks Mountains, and
- 2 Appalachian Mountains) and for four time periods (i.e., 1990-2018, 1990-1999, 2000-2009,
- 3 2010-2018).

6



## Figure 5A-54. Critical load uncertainty analysis for 14,943 values across the CONUS of the SSWC model. Blue and green dots have the lowest confidence interval and orange, and red dots have the highest confidence interval.

7 8 Critical loads used in the national assessment analysis used different methods (see 9 methods for more details). To understand differences in the CLs calculated with different 10 methods, waterbodies where methods overlap were compared. There are three main CL 11 approaches all based on watershed mass-balance approach where acid-base inputs are balanced. 12 The three approaches include: (1) SSWC model and F-Factor that is based on quantitative 13 relationships to water chemistry (Dupont et al. 2005, Scheffe et al. 2014, Lynch et al. 2022), (2) Statistical Regression Model that extrapolated weathering rates across the landscape using water 14 15 quality or landscape factors (Sullivan et al. 2012 and McDonnell et al. 2014), and (3) Dynamic 16 Models (MAGIC or Pnet-BGC). Critical load values were compared between these models to

17 determine model biases.

### 1 **5A.3.1 Results**

- 2 A Monte Carlo analysis was used to estimate the uncertainty around the CL. On average
- 3 the magnitude of the confidence interval for all SSWC CLs was 7.68 meq S/m<sup>2</sup>-yr or 1.3 Kg
- 4 S/ha/yr. The range based on the 5th to 95th magnitude of the confidence interval was 0.37-33.2
- 5 meq/m<sup>2</sup>/yr or 0.1-5.3 Kg S/ha/yr giving a confidence level of  $\pm 3.84$  meq/m<sup>2</sup>/yr or  $\pm 0.65$  Kg
- 6 S/ha/yr. Sixty-one percent of CL values had a low confidence level of less than 3.0325
- 7  $meq/m^2/yr$  or 0.5 Kg S/ha/yr, while 26% had levels greater than 6.25  $meq/m^2/yr$  or 1.0 Kg
- 8 S/ha/yr (Table 5A-49). Low confidence intervals were associated with CLs determined with
- 9 long-term water quality data and low variability in runoff measurements. CL values determined
- 10 by a single water quality measurement and in areas where runoff is variable (e.g. western U.S.)
- 11 had high uncertainty. CLs with the lowest uncertainty occurred in the eastern U.S., particularly
- 12 along the Appalachian Mountains, upper midwest, and Rockies Mountains (Figure 5A-55). Less
- 13 certain CLs were found in the midwest and south and along the CA to WA coast. Most of the
- 14 CLs in the midwest are based on a single or few water quality measurements while variability in
- 15 runoff in CA to WA coast account for those high uncertainty values.

### Table 5A-49. Results of the Monte Carlo analysis for uncertainty broken down by confidence interval.

Range of Confidence interval Kg/ha-yr	#. Values	Percent	
0.0 – 0.25	5462	37%	37%
0.25 – 0.5	3612	24%	61%
0.5 – 1.0	1994	13%	74%
1.0 – 2.0	903	6%	80%
>2.0	2972	20%	100%
Total	14943		

18 Table 5A-50 shows the average and 5<sup>th</sup> to 95<sup>th</sup> percentiles by ecoregions. Fifty-one

19 ecoregions had sufficient data to calculate the 5<sup>th</sup> to 95<sup>th</sup> percentile. Ecoregions in the

20 Appalachian Mountains on average (e.g. Northeastern Highlands (5.3.1), Blue Ridge (8.4.4),

21 Northern Lakes and Forests (5.2.1), and North Central Appalachians (5.3.3) and Rockies (e.g.

- 22 Sierra Nevada (6.2.14), Southern Rockies (6.2.14), and Idaho Batholith (6.2.15) had lower
- 23 uncertainty while Northeastern Coastal Zone (8.1.7), Cascades (6.2.7), Coast Range (7.1.8),
- 24 Interior Plateau (8.3.3), and Klamath Mountains/California High North Coast Range (6.2.11) had
- 25 on average high uncertainty.

Table 5A-50.	Results of the Monte Carlo analysis for uncertainty broken down by
	ecoregion. N/A indicates there was not sufficient data to calculate the
	percentile.

Ecoregio	on		Ave. 5 <sup>th</sup> – 95 <sup>th</sup> percentile		
Code	Name	No. Values	Kg S/ha/yr	meq/m²/yr	
5.3.1	Northeastern Highlands	2804	0.59 (0.05 - 2.07)	3.71 (0.32 - 12.96)	
8.4.4	Blue Ridge	2500	0.32 (0.06 - 0.9)	2 (0.39 - 5.61)	
8.4.1	Ridge and Valley	1497	1.64 (0.05 - 8.16)	10.25 (0.33 - 50.98)	
5.2.1	Northern Lakes and Forests	894	0.47 (0.02 - 2.04)	2.94 (0.12 - 12.76)	
8.3.4	Piedmont	573	1.29 (0.2 - 3.24)	8.09 (1.24 - 20.28)	
6.2.12	Sierra Nevada	566	0.41 (0.03 - 1.66)	2.57 (0.18 - 10.39)	
6.2.10	Middle Rockies	552	0.95 (0.08 - 5.08)	5.95 (0.53 - 31.76)	
6.2.14	Southern Rockies	444	0.58 (0.1 - 2.1)	3.62 (0.64 - 13.16)	
8.3.5	Southeastern Plains	413	1.59 (0.15 - 5.63)	9.94 (0.95 - 35.2)	
8.4.2	Central Appalachians	399	1.31 (0.08 - 3.4)	8.18 (0.47 - 21.27)	
8.1.8	Acadian Plains and Hills	371	1.2 (0.09 - 4.17)	7.47 (0.54 - 26.09)	
8.1.7	Northeastern Coastal Zone	323	2.38 (0.19 - 7.54)	14.87 (1.18 - 47.14)	
8.3.1	Northern Piedmont	265	4.1 (0.79 - 11.5)	25.6 (4.96 - 71.89)	
8.5.4	Atlantic Coastal Pine Barrens	233	1.1 (0.17 - 3.52)	6.87 (1.06 - 21.98)	
6.2.7	Cascades	229	3.68 (0.05 - 2.86)	22.97 (0.29 - 17.89)	
5.3.3	North Central Appalachians	222	0.6 (0.09 - 1.99)	3.75 (0.54 - 12.47)	
8.1.3	Northern Allegheny Plateau	217	1.46 (0.29 - 4.77)	9.11 (1.79 - 29.79)	
6.2.15	Idaho Batholith	212	0.51 (0.13 - 1.75)	3.21 (0.8 - 10.95)	
6.2.5	North Cascades	169	1.08 (0.15 - 4.73)	6.75 (0.96 - 29.55)	
8.3.7	South Central Plains	157	1.19 (0.32 - 3.09)	7.45 (2.03 - 19.34)	
8.5.3	Southern Coastal Plain	149	0.76 (0.1 - 2.89)	4.72 (0.6 - 18.09)	
8.4.9	Southwestern Appalachians	127	1.2 (0.18 - 4.71)	7.52 (1.15 - 29.46)	
7.1.8	Coast Range	119	5.88 (1.82 - 15.45)	36.77 (11.37 - 96.59)	
8.5.1	Middle Atlantic Coastal Plain	118	2.55 (0.26 - 9.04)	15.96 (1.63 - 56.48)	
6.2.13	Wasatch and Uinta Mountains	114	1.19 (0.15 - 7.11)	7.46 (0.95 - 44.44)	
8.1.4	North Central Hardwood Forests	101	2.3 (0.07 - 4.89)	14.4 (0.45 - 30.59)	
6.2.3	Northern Rockies	96	1.14 (0.19 - 4.84)	7.13 (1.18 - 30.27)	
8.3.3	Interior Plateau	89	5.44 (0.54 - 12.54)	34.01 (3.36 - 78.36)	
6.2.11	Klamath Mountains/California High North Coast Range	85	6.85 (0.43 - 18.46)	42.82 (2.67 - 115.34)	
8.1.1	Eastern Great Lakes Lowlands	72	2.69 (0.23 - 8.69)	16.83 (1.43 - 54.29)	
6.2.9	Blue Mountains	65	1.33 (0.26 - 4.22)	8.3 (1.62 - 26.37)	
8.4.5	Ozark Highlands	61	5.77 (1.22 - 9.5)	36.07 (7.6 - 59.39)	
8.4.8	Ouachita Mountains	51	0.94 (0.2 - 3.41)	5.88 (1.26 - 21.29)	
8.3.6	Mississippi Valley Loess Plains	41	3.1 (0.26 - 24.02)	19.39 (1.63 - 150.14)	
8.4.7	Arkansas Valley	39	1.31 (0.21 - 4.98)	8.2 (1.32 - 31.11)	

Ecoregio	n		Ave. 5 <sup>th</sup> – 95 <sup>th</sup> percentile	
Code	Name	No. Values	Kg S/ha/yr	meq/m²/yr
7.1.7	Puget Lowland	39	2.03 (0.29 - 5.77)	12.71 (1.81 - 36.08)
8.4.3	Western Allegheny Plateau	37	2.03 (0.41 - 4.89)	12.69 (2.55 - 30.55)
8.1.6	Southern Michigan/Northern Indiana Drift Plains	36	2.9 (0.75 - 5.21)	18.12 (4.66 - 32.56)
6.2.4	Canadian Rockies	32	2.5 (0.2 - 7.23)	15.59 (1.22 - 45.2)
6.2.8	Eastern Cascades Slopes and Foothills	32	1.52 (0.21 - 4.84)	9.51 (1.33 - 30.24)
9.4.5	Cross Timbers	31	3.72 (1.58 - 11.31)	23.24 (9.89 - 70.66)
9.2.3	Western Corn Belt Plains	27	3.91 (1.55 - 9.16)	24.43 (9.67 - 57.28)
13.1.1	Arizona/New Mexico Mountains	27	3.22 (0.28 - 10.53)	20.12 (1.74 - 65.79)
8.4.6	Boston Mountains	26	0.89 (0.23 - 4.12)	5.56 (1.42 - 25.72)
11.1.1	Central California Foothills and Coastal Mountains	25	10.79 (0.5 - 54.47)	67.41 (3.1 - 340.46)
9.2.4	Central Irregular Plains	24	3.08 (1.1 - 4.94)	19.27 (6.89 - 30.88)
7.1.9	Willamette Valley	24	3.43 (0.95 - 7.06)	21.45 (5.97 - 44.11)
11.1.3	Southern California Mountains	22	10.21 (1.5 - 20.12)	63.84 (9.4 - 125.78)
8.5.2	Mississippi Alluvial Plain	21	3.85 (0.95 - 9.94)	24.09 (5.91 - 62.1)
10.1.3	Northern Basin and Range	20	1.92 (0.35 - 8.81)	12.01 (2.18 - 55.05)
8.3.2	Interior River Valleys and Hills	19	4 (1.57 - 10.46)	25 (9.78 - 65.39)
10.1.5	Central Basin and Range	17	N/A	N/A
8.2.4	Eastern Corn Belt Plains	16	N/A	N/A
9.5.1	Western Gulf Coastal Plain	16	N/A	N/A
8.1.5	Driftless Area	15	N/A	N/A
8.1.10	Erie Drift Plain	14	N/A	N/A
8.3.8	East Central Texas Plains	11	N/A	N/A
8.2.1	Southeastern Wisconsin Till Plains	11	N/A	N/A
9.4.4	Flint Hills	9	N/A	N/A
9.4.2	Central Great Plains	5	N/A	N/A
10.1.4	Wyoming Basin	4	N/A	N/A
9.4.7	Texas Blackland Prairies	3	N/A	N/A
5.2.2	Northern Minnesota Wetlands	2	N/A	N/A
11.1.2	Central California Valley	2	N/A	N/A
10.1.8	Snake River Plain	2	N/A	N/A
10.1.2	Columbia Plateau	2	N/A	N/A
8.2.3	Central Corn Belt Plains	2	N/A	N/A
9.3.1	Northwestern Glaciated Plains	2	N/A	N/A
10.1.6	Colorado Plateaus	1	N/A	N/A

	Average (meq/m²/yr)	S.D. (meq/m²/yr)	5 <sup>th</sup> to 95 <sup>th</sup> (meq/m²/yr)	Magnitude & Confident Interval (meq/m²/yr)
New England Lakes				
All Years	0.7	1.05	0.01 – 2.87	0.15 (0.62 – 0.78)
1990 to 1999	0.8	1.17	0.00 – 3.10	0.30 (0.64 – 0.95)
2000 to 2009	0.92	1.18	0.01 – 3.88	0.29 (0.78 – 1.07)
2010 to 2018	0.36	0.59	0.01 – 1.48	0.15 (0.29 – 0.44)
Adirondacks Lakes				
All Years	8.82	7.79	0.13 – 23.52	0.77 (8.44 – 9.21)
1990 to 1999	11.71	9.01	0.72 – 27.83	1.62 (10.89 – 12.52)
2000 to 2009	9.28	7.11	0.68 – 21.2	1.16 (8.70 – 9.86)
2010 to 2018	5.73	6.01	0.00 – 17.91	1.03 (5.21 – 6.24)
Appalachian Streams				
All Years	3.27	5.77	0.03 - 13.68	0.52 (3.00-3.53)
1990 to 1999	5.05	7.29	0.43 - 20.18	1.14 (4.48 – 5.61)
2000 to 2009	2.43	4.75	0.00 - 11.61	0.73 (2.06 – 2.79)
2010 to 2018	2.27	4.30	0.00 - 10.82	0.70 (1.92 – 2.62)

1Table 5A-51.Results of the uncertainty analysis of Nitrate (NO3<sup>-</sup>) in EPA's Long-term2Monitoring (LTM) program.Unit are meq N/m<sup>2</sup>-yr.

4 The results of the uncertainty analysis of NO<sub>3</sub><sup>-</sup> flux (N leaching) based on the EPA's 5 LTM monitoring program are summarized in (Table 5A-51) by region and time period. Nitrate flux varied between regions with Adirondacks lakes having the highest annual fluxes and New 6 7 England Lakes with the lowest fluxes. Average values ranged from 0.36 to  $11.71 \text{ meq/m}^2/\text{yr}$  (0.5 8 to 1.6 Kg N/ha/yr). The ranges of confidence interval for the NO<sub>3</sub><sup>-</sup> flux differed some across the 9 monitoring sites from 0.15 to 1.62 meq/m<sup>2</sup>/yr (0.02 to 0.23 Kg N/ha/yr). A combined S and N confident interval was  $\pm 2.30$  to 3.77 meq/m<sup>2</sup>-yr which is equivalent to 0.37 to 0.60 Kg S/ha-yr 10 11 or 0.32 to 0.53 Kg N/ha-yr. While a comprehensive analysis of uncertainty has not been 12 completed for these data prior to the analysis include in this review, expert judgment suggested 13 the uncertainty for combined N and S CLs is on average about  $\pm 0.5$  kg/ha-yr (3.125 meg/m<sup>2</sup>/yr), which is consistent with the range of  $\pm 2.30$  to 3.77 meg/m<sup>2</sup>-yr determined from this analysis. 14 Given this consistency, an uncertainty of  $\pm 3.125 \text{ meg/m}^2$ -yr will be applied to the critical load 15 16 exceedances for the national, ecoregion, and case studies assessments. Watersheds determined 17 to exceed the critical load are those with exceedances above  $+3.125 \text{ meg/m}^2/\text{yr}$  while those that do not exceed will be below -3.125 meg/m<sup>2</sup>-yr. Those that fall between  $\pm 3.125$  meg/m<sup>2</sup>/yr will 18 be noted as "at the CL." 19 20 5A.3.1.1 **Critical Load Model Comparison** 

Results from the comparison between different CL methods are summarized below for
lakes in New England and the Adirondacks and streams in the Appalachian Mountains. For New

1 England and Adirondacks lakes, the MAGIC and the SSWC - F-Factor (Scheffe et al. 2014, Lynch et al. 2022) CL values were comparable with a R<sup>2</sup>=0.979 and R<sup>2</sup>=0.9587 and RMSE of 15 2 and 21 meq/m<sup>2</sup>/yr, respectively (Figure 5A-55). The Statistical Regression Model (Sullivan et al. 3 4 2014) CL estimates were also comparable to the SSWC - F-Factor model with a  $R^2 = 0.9815$ 5 (Figure 5A-56A). A bias towards higher values for the Statistical Regression Model (Sullivan et 6 al. 2014) was observed (Figure 5A-56B). However, this bias was not pronounced for CLs in the 7 range of 0 and 150 meq/m<sup>2</sup>/yr, where CL exceedance occur at current deposition levels. 8 For streams in the Appalachian Mountains, strong agreement was found between the 9 SSWC - F-Factor, Statistical Regression, and MAGIC models. McDonnell et al. (2014) found a highly correlated relationship ( $R^2 = 0.92$  and RMSE = 9-11 meq/m<sup>2</sup>/yr) between base cation 10 11 weathering estimates determined by MAGIC compared to the predictions based on weathering 12 rates using water quality or landscape factors. Additionally, CLs determined by MAGIC compared well to the SSWC - F-Factor were also highly correlated with a R<sup>2</sup>=0.9887 and RMSE 13 of 24 meq/m<sup>2</sup>/yr (Figure 5A-57A). However, the comparison was not as strong ( $R^2=0.8861$ ) 14 between CLs based on Statistical Regression Model (McDonnell et al. 2014) and the SSWC - F-15 16 Factor model (Scheffe et al. 2014, Lynch et al. 2022), indicating less agreement between those 17 methods (Figure 5A-57B). Overall, good agreement between the three methods used to calculate 18 CLs was found, indicating there was not a systematic bias between the methods and that they 19 should produce comparable results when used together. 20



Figure 5A-55. Critical load comparison between values based on MAGIC model (y-axis) and values based on the SSWC F-factor model (Lynch et al. 2022). Units are meq/m<sup>2</sup>/yr. a. New England Lakes and b. Adirondacks Lakes.



Figure 5A-56. Critical load comparison between values based on Regional Regression model (Sullivan et al. 2014) (y-axis) and values based on the SSWC F-factor model (Lynch et al. 2022). Units are meq/m<sup>2</sup>/yr. a. Full critical load range of comparison and b. Critical load range from 0 to 150 meq/m<sup>2</sup>/yr.



Figure 5A-57. A. Critical load comparison between values based on MAGIC model (y-axis) and values based on the SSWC F-factor model (Lynch et al. 2020) (x-axis). B. Critical load comparison between values based on Regional Regression model (Sullivan et al. 2014) (y-axis) and values based on the SSWC F-factor model (Lynch et al. 2022) (x-axis). Units are meq/m<sup>2</sup>/yr.

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### 1 5B.1 INTRODUCTION

2 This appendix summarizes salient aspects of key studies investigating responses of 3 terrestrial ecosystem components (trees, communities of herbs and shrubs, and lichens) to sulfur 4 and nitrogen deposition, and direct effects of the pollutants in ambient air. The effects may relate 5 to ecosystem acidification (e.g., acidification of soils in which plants are growing) or nutrient 6 enrichment (e.g., through changes in competitive advantages of nitrogen-limited species) or both. 7 The studies described here vary in the extent to which they clarify which factors may be eliciting 8 the responses. Two general types of studies are described in the sections that follow: controlled 9 addition experiments and observational (or gradient) studies. Each has strengths, limitations and 10 uncertainties associated with interpretation.

11 The strengths of the controlled addition study design include its ability to elucidate N- or 12 S-related factors and circumstances (e.g., chemical form, duration, concentration) that elicit a 13 response in the exposed plants (e.g., changes in growth rates of individual species, changes in 14 productivity of a forest plot, changes in community composition). The scope of impacts that can 15 be studied, however, is generally limited in the species included, and the size of terrestrial 16 community. Observational studies, in contrast, can include a large number and range of species 17 and terrestrial communities, including species less amenable to maintenance in controlled 18 experimental conditions. These studies, also called gradient studies as they provide for 19 consideration of observations across a gradient of pollutant concentrations, provide for the 20 assessment of numerous species and communities across large areas, including across 21 ecoregions.<sup>1</sup> Further, controlled addition studies, which generally include controls that have not 22 received additions, may be limited to assessment of responses to the addition of the specific 23 study chemicals. An observational study by its very nature involves the combined impact of 24 historical and contemporaneous atmospheric deposition in the study areas, which then poses 25 challenges to disentangling the effects of historic versus recent deposition and of the various chemicals deposited, as well as the effects of the soil chemistry and geology.<sup>2</sup> Further, the 26

<sup>&</sup>lt;sup>1</sup> Ecoregions are areas where ecosystems (and the type, quality, and quantity of environmental resources) are generally similar. The ecoregion framework referenced in this document is derived from Omernik (1987) and from mapping done in collaboration with EPA regional offices, other Federal agencies, state resource management agencies, and neighboring North American countries. Designed to serve as a spatial framework for ecosystems and ecosystem components, ecoregions denote areas of similarity in the mosaic of biotic, abiotic, terrestrial, and aquatic ecosystem components with humans being considered as part of the biota.

<sup>&</sup>lt;sup>2</sup> In context of 2015 ozone NAAQS review, and regarding potential use for predictive purposes in that review of a single-species O<sub>3</sub> gradient study involving tree seedlings planted in fields with transplanted soil at locations along a gradient in O<sub>3</sub> concentrations, CASAC, while noting it to provide important results, cautioned care in consideration for predictions in other circumstances of this single study that used a gradient methodology without experimental control of the pollutant exposures (Frey, 2014).

observational studies do not generally include measurements or assessments of the site soil
 chemistry or geology. Rather, they utilize atmospheric deposition estimates at assessment sites as
 surrogates for exposure conditions. These various strengths and limitations inform consideration
 of the studies below.

### 5 5B.2 TREE GROWTH AND SURVIVAL

6 As described in the ISA, acidic deposition, which can be comprised of S and N 7 compounds, can contribute to acidification of soils in which trees grow (ISA, section IS.5). 8 Deposition of N can also contribute to N enrichment of soil, which can increase the growth of N-9 limited trees. In a mixed forest, this can contribute to competitive advantages (depending on 10 species' growth rates), and potentially reducing the growth rate of out-competed species (ISA, 11 section IS.5.2). The relationship between deposition and these effects depends on soil status with 12 regard to acidification and N content, and accordingly is influenced by historic deposition and 13 the soil characteristics important to soil responses. As noted in the ISA, "[i]n areas where N and 14 S deposition has decreased, chemical recovery must first create physical and chemical conditions 15 favorable for growth, survival, and reproduction" for biological recovery to occur (ISA, p. IS-16 102). For example, although fewer studies have tracked potential recovery of terrestrial than 17 aquatic ecosystems, modeling studies in the southern Appalachian Mountains "suggest current 18 stress and recovery likely to take decades even under scenarios of large reductions in S 19 deposition" (ISA, p. 4-99). In the subsections below we provide details of several key studies in 20 the current ISA that evaluate relationships between N and S deposition on tree growth and 21 survival.

22

### 5B.2.1. Addition Studies

23 Several experiments involving S or N additions have been reported in the ISA focused on 24 study areas in the eastern U.S. These studies involve appreciable annual additions of S and/or N 25 compounds to experimental forest plots. While some study durations are limited to fewer than 26 five years, others extend appreciably longer than 10 years, providing the time to affect chemical 27 pools within the soil and the associated soil characteristics linked to acidification or nutrient 28 enrichment effects (e.g., Ca:Al ratio or NO<sub>3</sub> leaching). Among the studies summarized in Table 29 5B-1 below are addition studies that found species-specific results for growth and survival for 30 several eastern species including oaks, spruce, maples and pines. (Magill et al., 2004; McNulty et 31 al., 2005; Pregitzer et al., 2008; and Wallace et al., 2007). Further, some multiyear S/N addition 32 (>20 kg/ha-yr) experiments with small set of eastern species including sugar maple, aspen, white 33 spruce, yellow poplar, black cherry have not reported growth effects (Bethers et al., 2009; Moore 34 and Houle, 2013; Jung and Chang, 2012; Jensen et al., 2014).

5B-2

Location, Reference	Description	Additions	Tree specific Findings
Michigan (Pregitzer et al., 2008)	Four study areas across a 500 km gradient in temperature and N deposition in NW Michigan. Forests (approx 90 years old) dominated by sugar maple (82% by basal area). Study assessed soil biogeochemical properties, microbial communities, tree growth/ mortality.	30 kg N ha <sup>-1</sup> yr <sup>-1</sup> for 10 years starting in 1994 (as NaNO <sub>3</sub> ). Background deposition ranged from 6.8 to 11.8 kg N ha <sup>-1</sup> yr <sup>-1</sup> .	Increased growth (total live woody biomass) and mortality. Total deposition estimates: 36.8- 41.8 kg/ha-yr.
Mt. Ascutney, VT (McNulty et al., 2005)	Six study plots in montane spruce-fir forests. Assessed soil biogeochemical properties, microbial communities and tree growth and mortality.	15.7 and 31.4 kg N ha <sup>-1</sup> yr <sup>-1</sup> over 14 years starting in 1988 (as NH <sub>4</sub> Cl). Background deposition was 10 kg N ha <sup>-1</sup> yr <sup>-1</sup>	Reductions in total live basal area (low N-↓18%; high N ↓40% vs control↑9%), indicating reduced growth rates; increased red spruce mortality in high N.
Bear Brook, ME (Elvir et al., 2003; Bethers et al., 2009)	Two experimental watersheds (1 control and 1 treatment), each with softwood, mixed wood, and hard wood forest. Studies assessed soil biogeochemical properties, microbial communities and tree growth.	25.2 kg N ha <sup>-1</sup> yr <sup>-1</sup> and 28.8 kg S ha <sup>-1</sup> yr <sup>-1</sup> (as (NH <sub>4</sub> ) <sub>2</sub> SO <sub>4</sub> ) starting in 1989; assessed after 10 yrs. Initial background deposition was 8.4 kg N ha <sup>-1</sup> yr <sup>-1</sup> and 14.4 kg S ha <sup>-1</sup> yr <sup>-1</sup> .	Increased growth rates for sugar maple, but not for red spruce. No effect on sugar maple seedling density.
Northern Quebec, Canada (Houle and Moore, 2008; Moore and Houle, 2013)	Three year N addition (approximately 3x and 10x estimates of concurrent deposition), beginning in 2001, across 9 plots in black spruce and balsam fir boreal forests. Studies assessed NO <sub>3</sub> -leaching and tree growth.	9 and 30 kg N ha <sup>-1</sup> yr <sup>-1</sup> (spruce sites); 18 and 60 kg N ha <sup>-1</sup> yr <sup>-1</sup> (fir sites) (from ammonium nitrate additions) (as NH <sub>4</sub> NO <sub>3</sub> ) Background wet deposition of 3 kg N ha <sup>-1</sup> yr <sup>-1</sup> (black spruce forest) and 5.7 kg N ha <sup>-1</sup> yr <sup>-1</sup> (balsam fir)	After 3 years, no significant changes in growth rates for black spruce or balsam fir. After 8 years, no effect on sugar maple basal area growth.
Harvard Forest, MA (Magill et al., 2004)	Eight plots, four in a red pine plantation and four in a hardwood forest stand dominated by red and black oak, were assessed for tree growth and mortality.	50 and 150 kg N ha <sup>-1</sup> yr <sup>-1</sup> for 14 years starting in 1988 (as NH₄NO₃). Background deposition was 9 kg N ha <sup>-1</sup> yr <sup>-1</sup>	Increased growth (stand-level biomass), but no change in mortality in the hardwood forest. Decreased growth and increased mortality in the red pine plantation.
Canada Jung and Chang et al. (2012)	At study plots near Atasca oil sands, assessed above ground tree biomass. Main canopy species were quaking aspen and white spruce. Also included balsam fir, balsam poplar, black spruce and paper birch	30 kg N/ha-yr, 30 kg S /ha-yr and 30 kg N+30 kg S /ha-yr from 2006-2009	Biomass was increased in N-only treatment and was highest in the N+S treatment. Understory biomass unaffected. No evidence of increased NO <sub>3</sub> leaching
Millbrook, NY (Wallace et al., 2007)	Six pairs of plots in an upland mixed- oak forest dominated by chestnut oak, northern red oak and hickories at the Institute of Ecosystem Studies where studies assessed NO <sub>3</sub> - leaching, tree growth and mortality.	100 kg N ha <sup>-1</sup> yr <sup>-1</sup> (1996 to 1999), then 50 kg N ha <sup>-1</sup> yr <sup>-1</sup> (2000 to 2003) (as NH <sub>4</sub> NO <sub>3</sub> ). Background deposition was 10 kg N ha <sup>-1</sup> yr <sup>-1</sup>	Increased growth rates across species (oaks and hickories) and increased mortality in oaks.
Fenrow Forest, WV (May et al., 2005; Jensen et al., 2014)	Two paired watersheds, one control and one treatment. The most abundant species were red maple, tulip poplar and black cherry. Studies assessed soil biogeochemical properties and tree growth and mortality.	35 kg N ha <sup>-1</sup> yr <sup>-1</sup> and 40 kg S ha <sup>-1</sup> yr <sup>-1</sup> starting in 1989 (as (NH <sub>4</sub> ) <sub>2</sub> SO <sub>4</sub> ) Background deposition was approximately 15 kg N ha <sup>-1</sup> yr <sup>-1</sup> and 20 kg S ha <sup>-1</sup> yr <sup>-1</sup>	Reduced growth (stem diameter) in all 3 species (red maple, tulip poplar and black cherry) based on measurements taken in 1999 and 2001 (after 10 years of treatments). No difference in growth (basal area) for tulip poplar and black cherry after 22 yrs.

1	Table 5B-1.	<b>Experimental additio</b>	n studies assessing	tree growth	and/or survival.
1		Experimental auditio	n studies assessing	u ce gi omu	and/or survival.
### 5B.2.2. Gradient or Observational Studies

2 The evidence newly available in this review includes observational or gradient studies 3 that investigated the existence of statistically significant associations of tree growth and survival 4 or mortality with S or N deposition (Table 5B-2; ISA, Appendix 5, section 5.5.2 and Appendix 6, 5 sections 6.2.3.1, 6.3.3 and 6.6.1). In general, these studies utilized measurements of tree growth 6 and survival or mortality across multiyear intervals at designated plots, and estimates of average 7 S and/or N deposition (or in some cases, emissions estimates) in the same locations. Statistical 8 models were employed in the analyses, and the influence of different sets of additional factors 9 (e.g., related to climate, other air pollutants, topography and stand characteristics).

Table 5B-2 below summarizes these studies some of which focused on regions within a state and others which encompassed multistate regions. The three larger studies utilized data from the USFS Forest Inventory and Analysis (FIA) program in which measurements are taken at approximately 5 year intervals at designated plots in forests across the U.S. The three studies have utilized USFS-FIA data for different, but overlapping study areas (Figure 5B-1) and

15 species. More detailed descriptions of these studies and their findings are provided in sections

16 5B.2.2.1 through 5B.2.2.3 below.



Figure 5B-1. Study areas of three observational studies utilizing FIA plot data. The western
 extent of Dietze and Moorcroft (2011) is a rough approximation.

- 1 Other observational studies in the recently available evidence have investigated
- 2 relationships of tree growth with estimates of SOx and NOx emissions. For example, increases in
- 3 eastern redcedar growth in West Virginia has been associated with reductions in SO<sub>2</sub> emissions
- 4 and increases in atmospheric CO<sub>2</sub> concentrations (Thomas et al., 2013). In a North Carolina
- 5 high-elevation forest, increases in red spruce radial growth since the late 1980s has been
- 6 associated with declining SOx and NOx emissions from SE utilities, as well as increasing
- 7 temperatures and CO<sub>2</sub> (Soule, 2011). Recent studies in areas of Europe where SO<sub>2</sub>
- 8 concentrations are generally higher than in the U.S. have also reported increased growth of some
- 9 conifer species (e.g., silver fir) to be related to reductions in SO<sub>2</sub> concentrations (ISA, Appendix
- 10 3, section 3.2).
- 11

Table 5B-2. Recent gradient/observational studies of associations between tree growth and
survival or mortality and S or N deposition: smaller-scale studies.

Study	Description	Summary					
Smaller Regional Scales							
Bedison and McNeil 2009	32 plots in northern hardwood and subalpine spruce-fir dominated forest plots in Adirondack Park, NY. Trees were measured in1984 and 2004. The spatial pattern of inorganic N deposition in wet deposition was estimated across the plot locations by multiple regression. Analyses performed for growth of both individual species and all individuals within each plot. Potential influence of S deposition was not assessed.	At the species level, positive associations of growth with N deposition were found for maple, spruce and fir species, with the largest growth increases in red maple, balsam fir and red spruce. Responses varied by forest type and size class.					
Sullivan et al 2013	Study focused on 50 plots in western Adirondack region with commonly occurring sugar maple and a 10-fold range of Ca availability (based on previous stream and soil studies). Plant measures included DBH of all trees > 10 cm within plots, assessment of sugar maple canopy condition and vigor, dendrochronology of sugar maple trees, and seedling and sappling counts in subplots. Soil chemistry measurements included base saturation, exchangeable calcium, exchangeable magnesium and soil pH. Total S and inorganic (nitrate and ammonium) N deposition estimated using empirically based GIS model. Average annual (based on the period 2000-2004) N deposition was calculated as the product of estimated average annual precipitation from PRISM5, based on 30-year normals (1970-2000) and kriged S and N precipitation chemistry from NADP locations. Dry deposition of SO <sub>4</sub> -S, HNO <sub>3</sub> -N, and particulate NO <sub>3</sub> -N and NH <sub>4</sub> -N across Adirondack region calculated as products of air concentrations, based on the average of 2000-2004 CASTNet air chemistry data, and vegetation cover deposition velocities per CASTNet protocols.	<ul> <li>Plots with lower soil base saturation did not have sugar maple regeneration, with the proportion of sugar maple seedlings dropping off substantially from at/above approximately 60% for base saturation levels at/above 20% to at/below approximately 20% for base saturation at/below about 10%.</li> <li>Canopy vigor was positively correlated with soil pH and exchangeable Ca, Mg.</li> <li>Mean growth rates (BAI) were positively correlated with exchangeable Ca and base saturation at the watershed level. Sugar maple distribution negatively associated with estimated average 2000-04 N+S deposition (750-1120 eq/ha/yr)</li> </ul>					

1	Table 5B-3.	Recent gradient/observational studies of associations between tree growth and
2		survival or mortality and S or N deposition: larger-scale FIA data studies.

Study	Description	Summary					
	Larger Regional and National Scales (and using USFS FIA data)						
Thomas et al 2010	Assessed 24 of the most common northeastern tree species using 20,067 FIA plots in 19 states from 1978 to 2001, with the measurement interval varying from 8.3 to 14.4 across states. Tree growth and survival were assessed with regard to association with N deposition (mean annual total N deposition, 2000-04)	Growth of 11 species was positively associated with N deposition (including all species with arbuscular mycorrhizal fungi associations). Growth of 3 species was negatively associated with N deposition. Survival of 8 species was negatively associated with N deposition, with positive associations for 3 species.					
Dietze and Moorcroft	Assessed influence of patterns of $SO_4^-$ and $NO_3^-$ wet deposition (1994-2005 average), $O_3$ (1996- 2006) and climate, topographic and tree stand	Mortality in 7 of the functional groups was positively associated with both $SO_4^-$ and $O_3$ ; and negatively associated in 1 group.					
2011	factors) on observed variation in tree mortality at FIA plots in the eastern and central U.S. from 1971 to 2005, binning the 267 species into 10 plant functional types.	Mortality in 9 of the 10 functional groups was negatively associated with NO <sub>3</sub> , and positively associated in 1 functional group.					
Horn et al 2018	At USFS/FIA plots across the continental U.S., analyzed potential for associations of growth and	Growth in 31 species was negatively associated with S deposition.					
	generally 10 years) with estimates of average N and S deposition for the same interval, all within	Survival in 40 species was negatively associated with S deposition.					
	the period, 2000-2013. Other factors included in the analysis were temperature, precipitation, and terms representing the influence of tree size and competition on growth and survival. Deposition estimates were drawn from TDEP dataset of NADP's Science Committee on Total Deposition for the measurement interval of each plot. The analyses focused on 71 species that met criteria for sample size (>2000 trees for both growth and survival datasets) and for collinearity (correlation among the independent variables) of N or S, separately, with the other three independent variables (S or N, temperature and precipitation) for growth or survival (Variance Inflation Factor < 3).	Growth in 20 species was positively associated with N deposition and in 2 species (yellow birch and eastern hemlock) was negatively associated. Growth in 17 other species was positively associated with N deposition at lower levels and negatively associated at higher levels. Survival of 3 species was positively associated with N deposition and in 6 species was negatively associated. Survival in 25 other species was positively associated with N deposition at lower N deposition and negatively associated at higher levels.					

4

## 5B.2.2.1. Dietze and Moorcroft (2011)

5 The study by Dietze and Moorcroft (2011) statistically analyzed patterns of tree mortality 6 in the eastern and central U.S. using FIA data from 1971 to 2005. The total sample size was 3.4 7 million tree measurements and 750,000 plot level measurements. Mortality was quantified as a 8 binary metric (lived or died) based on resampling of FIA plots after intervals of 5 to 15 years.

1 Climate data were extracted from the database maintained by the PRISM database.<sup>3</sup> Using data

- 2 from 1971 to 2000 the annual average precipitation, average monthly minimum temperature
- 3 across December, January and February, and the average monthly maximum temperature across
- 4 June, July and August were calculated. Air quality data were obtained from the National
- 5 Atmospheric Deposition Program (NADP) for estimates of wet deposition (in kg ha<sup>-1</sup>yr<sup>-1</sup>) for
- 6 ammonium (NH<sub>4</sub><sup>+</sup>), nitrate (NO<sub>3</sub><sup>-</sup>), hydrogen ion (H<sup>+</sup>) and sulfate (SO<sub>4</sub><sup>2-</sup>) for the period of 1994-
- 7 2005 and from the EPA's AIRDATA database for ozone for the period of 1996-2006. The ranges
- 8 of sulfate and nitrate wet deposition estimates across the study area were 4 to 30 kg/ha-yr and 6
- 9 to 16 kg/ha-yr, respectively (Dietze and Moorcroft, 2011). There were 267 tree species sampled
- 10 in the study region. The species were classified into 10 different plant functional types to
- 11 facilitate analyses (see Attachment 1). The mortality analysis utilized a logistic regression model
- 12 for binary mortality probability, relating the mortality probability (live or dead) to a linear model
- 13 of the covariates.
- 14 All 13 covariates<sup>4</sup> were found to be statistically significant predictors of mortality for
- 15 more than one of the plant functional types. Sulfate deposition demonstrated a significant
- 16 positive effect on mortality in seven of the 10 plant functional groups and a slight negative effect
- 17 in one group (Table 5B-4). Nitrate deposition demonstrated a significant negative effect on
- 18 mortality in 9 of the 10 plant functional groups and a positive effect in the tenth.
- 19 Of note is that ozone exhibited the same pattern of effects as  $SO_4^-$  (Table 5B-4). The
- 20 authors also noted correlations between the nitrate and sulfate wet deposition estimates
- 21 (correlation coefficient of 0.82), and that the highest deposition estimates were for the Ohio
- 22 River valley and the northeastern United States (Dietze and Moorcroft, 2011).
- 23

<sup>&</sup>lt;sup>3</sup> The PRISM (Parameter-elevation Regressions on Independent Slopes Model) database is maintained by the PRISM Climate Group who compile data from multiple monitoring networks and develop spatial climate datasets to investigate short- and long-term climate patterns. https://prism.oregonstate.edu/

<sup>&</sup>lt;sup>4</sup> There were 13 covariates in 4 categories: climate (mean annual precipitation, mean summer maximum temperature, mean winter temperature), air pollutants (NO<sub>3</sub><sup>-</sup>, SO<sub>4</sub><sup>2-</sup>, O<sub>3</sub>), topography (topographic convergence index, elevation, slope, radiation index), and stand characteristics (stand basal area, stand age, and focal tree DBH).

#### 1 Table 5B-4. Influence of three air pollutants on pattern of tree mortality for 10 plant 2 functional groups in the eastern and central U.S. (drawn from Dietze and 3 Moorcroft, 2011).

Plant Functional Group	Sulfate, wet deposition	Nitrate, wet deposition	Ozone
Early Successional. Hardwood	Pos	Neg	Pos
Evergreen Hardwood	Pos	Neg	Pos
Hydric	Pos	Neg	Pos
Late Successional Conifer	Neg	Neg	Neg
Late Successional Hardwood	Pos	Neg	Pos
Midsuccessional Conifer		Neg	
Northern Midsuccessional Hardwood		Pos	
Northern Pine	Pos	Neg	Pos
Southern Midsuccessional Hardwood	Pos	Neg	Pos
Southern Pine	Pos	Neg	Pos

In this study, which was limited to the eastern and central U.S., the deposition metrics 4 were based on wet deposition estimates for  $SO_4^{2^-}$ , as an indicator of acid deposition,<sup>5</sup> and  $NO_3^{-}$ , 5 as an indicator of wet deposition of total N (Dietze and Moorcroft, 2011).<sup>6</sup> As noted by the 6 7 authors, "[t]he impacts of both acidification and nitrogen deposition on tree mortality result from 8 cumulative, long-term deposition, and the patterns presented here should be interpreted in that 9 light," further noting that "these relationships are not intended to assess the impacts of interannual variability in deposition nor the efficacy of NO<sub>3</sub><sup>-</sup> or SO<sub>4</sub><sup>2-</sup> regulation" (Dietze and 10 11 Moorcroft, 2011). Different patterns and associations might be found for analyses utilizing total 12 deposition (wet and dry) and for species and locations in the western U.S., with its differing 13 species, soils, climate and historic deposition patterns. In order to utilize all the measurements, 14 including those for species with lower sample sizes, the tree species were categorized into plant 15 functional groups; accordingly, variation in mortality at species level was not assessed. 16 5B.2.2.2.

#### **Thomas et al. (2010)**

17 The study by Thomas et al. (2010) statistically analyzed relationships of growth and

18 survival to N deposition for 24 commonly occurring tree species in a 19-state region of the U.S.

19 The study region included USFS FIA program plots in 19 states, bounded by Maine in the

- 20 Northeast to Virginia and Kentucky in the South, and west to Wisconsin and Illinois. Data were
- 21 extracted for the 24 tree species at 20,067 plots. Two measurements were taken at these plots

<sup>&</sup>lt;sup>5</sup> Preliminary analyses indicated stronger relationship for tree mortality with SO<sub>4</sub> than with hydrogen ion (Dietze and Moorcroft, 2011).

<sup>&</sup>lt;sup>6</sup> Preliminary analyses indicated a stronger relationship for tree mortality with NO<sub>3</sub> than with NH<sub>4</sub> or total N (Dietze and Moorcroft, 2011).

during the period from the 1978 to 2001, with the measurement interval varying across the 19
 states from 8.3 to 14.4 years (Thomas et al., 2010, supplemental information).

Nitrogen deposition was estimated using NADP wet deposition estimates and CASTNET
dry deposition estimates for the period from 2000 through 2004. Total N deposition estimates at
the study plots for this period ranged from 3 to 11 kg N/ha-yr (Thomas et al 2010, Supplemental
Information). Precipitation and temperature were calculated from PRISM with plot specific

7 values for the span of years from first measurement to second measurement.

8 The statistical analyses tested a suite of alternate regression models for growth and 9 survival response to N deposition, precipitation and temperature. Additional ecological attributes 10 included in the study, but not in the models relating growth or survival to the N deposition 11 metric, were plant functional type (deciduous hardwood or evergreen conifer) and mycorrhizal 12 fungi association (arbuscular versus ectomycorrhizal). The Akaike Information Criteria (AIC) 13 was used to select the most parsimonious model (i.e., the best model fit for the fewest 14 parameters).

15 Variation in tree growth for 14 of the 24 species was found to be significantly associated 16 with N deposition, with positive associations (greater growth at sites with greater N deposition) 17 found for 11 species and negative associations for three species. All three species with negative 18 associations were evergreen conifers (red pine, red spruce, and white cedar) that varied widely in 19 the amount of growth variation per kg N/ha-yr from -9% for red pine to -0.1% and -0.01% for 20 the other two species, respectively (Thomas et al., 2010). Three of the four most abundant 21 species (red maple, sugar maple and northern red oak) exhibited strong positive associations. The 22 largest variation in growth per unit variation in the N deposition metric was observed for black 23 cherry, tulip poplar, scarlet oak, white ash and balsam fir (18 to 12.3% difference in growth per 24 kg N/ha-yr).

With regard to probability of tree survival, variation in survival probability across the 25 26 study area was significantly associated with the N deposition metric for 11 of 24 species 27 examined. The association was negative for eight species, with the largest survival variation per 28 kg N/ha-yr observed for scarlet oak (-1.67%) and quaking aspen (-1.3%). The association was 29 positive for three species (red maple, paper birch, and black cherry), with only one of the three 30 having a survival variation per kg N/ha-yr above 1%, black cherry (Thomas et al., 2010). 31 The authors also suggest that the type of mycorrhizal fungi association with the tree 32 species may influence its response to N deposition as all five species with arbuscular mycorrhizal 33 fungi associations had positive associations of growth with N deposition and all 8 of the species 34 with negative associations of survival with N deposition had ectomycorrhizal fungi associations 35 (Thomas et al., 2010). Mycorrhizal fungi are important for supplying nutrients and water to 36 plants, influencing soil C sequestration, and producing mushrooms (ISA, p. ES-16). Mycorrhizal

1 fungi have long been observed to be sensitive to increased forest N availability (ISA, Appendix

2 6, section 6.2.3.2).

Not included in the analysis were several factors with the potential to influence tree growth and survival, including competition, soil chemistry, S deposition and ozone. Accordingly, there was also no analysis of collinearity between such parameters. Most notably, there was no assessment of the extent of N deposition correlation with S deposition and/or ozone. The study area and species list was the most limited of the three observational studies relying on USFS-FIA data.

9

#### 5B.2.2.3. Horn et al. (2018)

10 The most recent analysis utilizing the USFS-FIA data, by Horn et al. (2018) also covers 11 the largest area. This study relies on tree measurements taken for approximately 1.4 million trees 12 across approximately 70,000 FIA plots. The plots are scattered across 47 states of the contiguous 13 U.S., excluding Wyoming<sup>7</sup> (Figure 5B-2; Horn et al., 2018, Supplemental Data). The eastern 14 U.S. has many more plots than the West and the areas with highest densities of plots (and 15 associated measurements) include Wisconsin, northern Michigan and Minnesota and New 16 England (Figure 5B-2).<sup>8</sup> 17 The study was similar in approach to Thomas et al. (2010), and investigated associations 18 between variation in tree growth and survival and atmospheric deposition of N and S across the 19 plots for each species. The tree growth and survival measurements were those collected by the 20 FIA generally within the years from 2000 to 2016, with the remeasurement interval for each plot varying by state and inventory cycle from 8.8 to 12.1 years (Horn et al., 2018, Supplemental 21 22 Data). The most common measurement interval across all plots in the study dataset was 10 years 23 (Horn et al., 2018).

<sup>&</sup>lt;sup>7</sup> The lack of plots in Wyoming resulted because when the researchers obtained the FIA in January 2017, although there were FIA plots in Wyoming, there were no re-measured plots which is a requirement to assess rates of growth and survival.

<sup>&</sup>lt;sup>8</sup> This observation is the result of there being more plots in the eastern US due to greater forested area. Within all U.S. forested areas, plot density is the same by the FIA design (Bechteld and Patterson, 2005).



#### 2 Figure 5B-2. Location of FIA plots, based on survival analysis of Horn et al. (2018).

3 Individual tree data were available for a total of 151 species, with 94 species meeting the 4 study threshold of 2000 individual trees for both growth and survival data (Horn et al., 2018). 5 Tree growth values were in terms of biomass gains based on measurements of individual trees at 6 the USFS FIA plots during initial and follow-up visits. Survival was assessed by observing 7 whether a tree observed on an initial visit was still alive at the follow-up visit (e.g., survived or 8 not). Thus, survival is a probability metric of the tree surviving and the relationship of survival 9 (y/n) with the average deposition at that site across years between visits was statistically 10 analyzed (along with other co-factors like temperature, precipitation, size, competition, and N or 11 S deposition). 12 The N and S deposition estimates for each plot's measurement interval were derived from 13 spatially modeled N and S deposition estimates available from the U.S. National Atmospheric

14 Deposition Program's Total Deposition Science Committee (stored on the U.S. EPA's FTP

- 15 server). Average N deposition and S deposition for each plot were derived from the annual
- 16 deposition estimates for the years included in the measurement interval (from year of first
- 17 measurement to year of follow-up measurement) for the plot. The plot-level deposition estimates

5B-12

1 were assigned to all the trees in that plot. Temperature and precipitation values were obtained 2 from PRISM Climate Group<sup>9</sup> and assigned to individual plot values, as for N and S deposition. 3 In addition to temperature and precipitation, other parameters analyzed in the statistical 4 models included tree size, and competition. A total of 5 different models of growth as a function 5 of various sets of the 7 parameters were examined: 1) a full model with the size, competition, 6 climate, S deposition, and N deposition terms; 2) a model with all terms except the N deposition 7 term; 3) a model with all terms except the S deposition term; 4) a model with all terms but 8 without S and N deposition terms; and 5) a null model that estimated a single parameter for the 9 mean growth parameter. For survival, a total of 9 different models were examined, the same 5 as 10 for growth plus additional models using 2 different size estimates. S deposition was constrained 11 to have a flat or decreasing response while N deposition could have flat, increasing or decreasing 12 effects. The models selected to describe growth and survival for each species were the simplest 13 models (i.e., the one with the fewest parameters) that were within 2.0 AIC units of the best 14 model (i.e. the model with the lowest AIC) following Thomas et al. (2010). 15 To quantify collinearity of N and S deposition against other environmental variables, the 16 study calculated variance inflation factors (VIF). This was done for each tree species and for

16 study calculated variance inflation factors (VIF). This was done for each tree species and for 17 both growth and survival. While VIF values above 10 have been presented in the literature as a 18 threshold for high collinearity, the authors used VIF < 3 as a criterion for species inclusion to be 19 conservative (Horn et al., 2018). The growth and/or survival models for 71 of the 94 species

20 analyzed met this criterion. Although not utilized in model assessments for each species,

correlation coefficients were calculated for N and S deposition across the plots assessed for that
 species (Horn et al., 2018, supplemental information).

23 Of the 71 species, growth of 31 and survival probability for 40 were negatively 24 associated with the S deposition metric values. For 21 species, both growth and survival were 25 negatively associated with S deposition. No statistically significant association was observed for 26 growth or survival in 5 of the 71 species (Horn et al., 2018). With regard to N, among the 27 statistically significant models for growth and survival for some species were hump-shaped 28 relationships, with positive associations in the lower part of the range of N deposition estimates 29 for a species and negative associations in the upper part of the range. This was the case for 30 growth and N deposition for 17 species and for survival of 25 species. Growth for two species 31 and survival for six was negatively associated with the N deposition metric across their ranges. 32 Conversely, positive associations across the full range were found for growth of 20 species and

<sup>&</sup>lt;sup>9</sup> The PRISM climate group at Oregon State University, supported by the USDA, collect climate data and apply modeling techniques to develop publicly available datasets covering the period from 1895 to the present. The Parameter-elevation Relationships on Independent Slopes Model (PRISM) is an interpolation method used in developing the data. (PRISM Climate Group, Oregon State University, https://prism.oregonstate.edu).

survival of one species, black locust (Robinia pseudoacacia),<sup>10</sup> which was also among those with
 positive growth associations.

3 Multiple factors with potential impacts on tree growth/survival were not assessed, 4 including ozone and others, such as disturbance history (Latty et al., 2003) and insect infestation 5 (Eshleman et al., 1998, 2004). Further, the influence of soil characteristics on growth or survival 6 was also not analyzed. However, so long as these other factors do not spatially and temporally 7 correlate with N or S deposition, the omission of these factors would not affect the reported 8 relationships. Significantly, the study does not account for the influence at the FIA plots of 9 higher historical deposition. So the extent to which observed associations relate to historically 10 higher deposition is unclear. Thus, the extent to which relationships reported for N and S 11 deposition could have had unaccounted for influences of these variables and associated impacts 12 is unknown. 13 The authors express strongest confidence in findings from this gradient analysis for the 14 Eastern U.S., noting the smaller gradients in deposition and smaller number of different species 15 at western plots (Horn et al 2018). This uncertainty in the west primarily had to do with the 16 sulfur relationships, which were based on often shorter S deposition gradients that were often

- 17 highly correlated with N despite lower VIF scores overall. Plots for some species (e.g., Utah
- 18 juniper, Douglas fir) were only in the West (Table 5B-5), FIA plots for some other species are
- 19 predominantly in the Eastern U.S. (northeast, mid-atlantic or south), or in the Midwest (e.g.,
- 20 upper Great Lakes areas). Given the lesser confidence for species only at western plots, we have
- 21 focused discussion below on the species for which the sample sites were not limited to the
- 22 western U.S.

# Table 5B-5. Species with significant growth or survival associations with S or N deposition for which FIA sites are only in western states (drawn from Horn et al., 2018).

All FIA assessment sites in western states				
Genus species	Common name			
Juniperus osteosperma	Utah juniper			
Lithocarpus densiflorus	Tanoak			
Pinus monophyla	Singleleaf pinyon			
Pseudotsuga menziesii	Douglas fir			
Tsuga heterophylla	Western hemlock			

25 Examination of the correlation coefficients additionally indicates relatively high N/S

26 correlations for some species, complicating interpretation. For example, across the 71 species,

<sup>&</sup>lt;sup>10</sup> More than 90% of sample sites for this species were in ecoregions 8.1 – 8.4, with more than 50% in 8.4 (Ozark, Ouachita-Appalachian Forests), regions heavily impacted by SO<sub>2</sub> and acid deposition in the past (ISA, Figure 2-70); the N/S correlation coefficient for these sampling sites was 0.18 (Horn et al., 2018, Supplemental Figures).

- 1 the two highest correlation coefficients are those for eastern hemlock (0.78) and American beech
- 2 (0.76), and four of the six species with the next highest coefficients are also for species whose
- 3 ranges are concentrated in the eastern U.S. (pond cypress [0.71], yellow birch [0.7], sugar maple
- 4 [0.67], pitch pine [0.66]) (Horn et al., 2018, supplemental information). Differences in
- 5 quantitative relationships among species may reflect, in part, differences in geographic
- 6 distribution of sampling locations, with some species' sites largely concentrated in just a couple
- 7 of ecoregions (e.g., paper birch in the far north Great Lakes and Appalachians). Thus, differences
- 8 in geographic distributions of the species contribute to differences in ranges of deposition
- 9 history, geochemistry, etc, and may contribute to findings reported for some species.
- 10 Across sites of species with statistically significant associations of growth or survival
- 11 with the S deposition metric, the median average measurement-interval S deposition value,<sup>11</sup>
- 12 with a few exceptions ranged from 5 to  $12 \text{ kg S ha}^{-1} \text{yr}^{-1}$ . Focusing first on association for growth,
- 13 the median S deposition metric values for the species for which growth was negatively
- 14 associated with S dep (excluding the two species with samples only in the west) ranged from 4 to
- 15 12 kg S ha<sup>-1</sup>yr<sup>-1</sup>, with values below 5 kg S ha<sup>-1</sup>yr<sup>-1</sup> for two species, paper birch and white spruce
- 16 (for which 75-80% of sites were in the Northern Forests ecoregion<sup>12</sup>) and above 10 kg S ha<sup>-1</sup>yr<sup>-1</sup>
- 17 for two species, black locust and sweet birch, which have 70% to more than 90% of their sites in
- 18 the Eastern Temperate Forests ecoregion<sup>13</sup> (Figure 5B-3; distribution of measurement sites
- 19 shown in Attachment 2A).

<sup>&</sup>lt;sup>11</sup> Median average measurement-interval S and N deposition values cited in this document are rounded to whole numbers.

<sup>&</sup>lt;sup>12</sup> The Northern Forests is the level 1 ecoregion (5.0), which in the U.S. is located in northern Michigan, Wisconsin and Minnesota (https://www.epa.gov/eco-research/ecoregions-north-america).

<sup>&</sup>lt;sup>13</sup> Eastern Temperate Forests is the level 1 ecoregion (5.0), which includes most of the eastern U.S. (https://www.epa.gov/eco-research/ecoregions-north-america).



# Figure 5B-3. Average measurement interval S deposition at sites of species with negative growth associations with S deposition metric (drawn from Horn et al., 2018).

The median deposition metric values for the 40 species for which survival probability was negatively associated with S deposition ranged from 3 to 12 kg S ha ha<sup>-1</sup>yr<sup>-1</sup> (Figure 5B-4). Values for ten species were at or above 10 and for two were below 5 kg S ha ha<sup>-1</sup>yr<sup>-1</sup>. The two values below 5 were for paper birch, for which nearly 80% of the measurement sites were in the Northern Forests ecoregion, and quaking aspen, for which more than 60% of the sites were in the Northern Forests ecoregion and another 16% were in the Southern Rockies and Wasatch and Uinta Mountains (see sample distribution in Attachment).



# Figure 5B-4. Average measurement-interval S deposition at sites of species with negative survival associations with S deposition metric (drawn from Horn et al., 2018).

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1 With regard to N deposition, of the 39 species with significant associations of growth

2 with N deposition, the association was negative across the full deposition range of their sites for

- 3 two species, pitch pine and bur oak. These species' sites were predominantly in the Atlantic
- 4 coastal pine barrens and northern plains and forests, respectively. The median deposition across
- 5 all sites of these species were 9 and 10 kg N ha  $ha^{-1}yr^{-1}$  (Figure 5B-5). The median deposition
- 6 values for the two other species, with hump shaped functions that were negative at the median,<sup>14</sup>
- 7 were 7 and 8 kg N ha ha<sup>-1</sup>yr<sup>-1</sup>, respectively (Figure 5B-5).



8

Figure 5B-5. Average measurement-interval deposition at sites of species with negative
 associations of growth with N deposition metric at median (drawn from Horn
 et al., 2018). Blue asterisks indicate species with hump shape associations.

12 Of the remaining 35 species with significant associations of growth with measurement-13 interval N deposition, the association was positive across the full deposition range of their sites

- 14 for 20 species. The median N deposition metric values for the 17 nonwestern species<sup>15</sup> of these
- 15 20 species ranged from 7 kg N ha ha<sup>-1</sup>yr<sup>-1</sup> (for a number of species) up to 12 kg N ha ha<sup>-1</sup>yr<sup>-1</sup> for
- 16 silver maple, hackberry and black walnut (Figure 5B-6). For the 15 species with significant
- 17 associations of growth with measurement interval N deposition that were positive at the median
- 18 average measurement-interval deposition for the species, one was a western species, western

<sup>&</sup>lt;sup>14</sup> Given its role as a measure of central tendency of a dataset, the nature of the association for hump shape models at the median is what is described in the groupings here.

<sup>&</sup>lt;sup>15</sup> Three western species, Utah juniper, Douglas fir and western hemlock (Table 5B-5) had positive growth association across range of N deposition metric values.

- 1 hemlock (Table 5B-5). The median average measurement-interval deposition metric values for
- 2 the other 14 species ranged from 7 to 11 kg N ha  $ha^{-1}yr^{-1}$  (Figure 5B-6).



# Figure 5B-6. Average measurement-interval deposition at sites of species with positive associations of growth with N deposition metric at median (drawn from Horn et al., 2018). Blue asterisks indicate species with hump shape associations.

7 Of the six species with negative associations of survival with the N deposition metric 8 across the full range of the N deposition metric (water oak, southern red oak, winged elm, scarlet 9 oak, mockernut hickory and American elm), the median deposition values ranged from 8 to 11 kg N ha ha<sup>-1</sup>yr<sup>-1</sup>.(Figure 5B-7). The median deposition values for all of the 21 other species with 10 11 hump shape functions that were negative at the median deposition value ranged from 3 to 11 kg N ha ha<sup>-1</sup>yr<sup>-1</sup> (Figure 5B-7; see blue asterisks). The values for the 19 species for which sample 12 sites were not limited to the western U.S. ranged from 7 to 12 kg N ha ha<sup>-1</sup>yr<sup>-1</sup>. The four values 13 14 below 9 were for quaking aspen (75% sites in Northern Forests, Wasatch and Uinta Mtns and 15 Southern Rockies), slash pine (~60% sites in southern coastal plain), eastern hemlock (~50% 16 sites in Northern Forests and ~30% in Mixed Wood Plains) and red pine (nearly 70% in Northern 17 Forests).



# associations of survival with N deposition metric (drawn from Horn et al., 2018). Blue asterisks indicate species with hump shape associations.

5 Turning to positive associations of survival with N, there was 1 species (black locust) 6 with a positive associations of survival with N across the full deposition range with a median 7 deposition of 11 kg N ha ha<sup>-1</sup>yr<sup>-1</sup> (Figure 5B-8). The median deposition values for the 4 species 8 with hump-shaped associations that were positive at the median ranged from 7 to 12 kg N ha ha<sup>-1</sup> 9  $^{1}$ yr<sup>-1</sup>. The two values below 10 were for paper birch, for which nearly 80% of the measurement 10 sites were in the Northern Forests ecoregion, and American beech with more than 50% of sites in 11 Northern Forests (and N/S correlation coefficient of 0.76).

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1			*	*			
2	Figure 5B-8. Average	measurement-	interval d	leposition	at sites of	f species w	ith positive
3 1	associatio 2018) Bh	ons of survival	with N d	eposition r	netric (di	rawn iron	i Horn et al.,
-	2010). Di		ieute spee		mp snape		113.
5							
6 7	5B.2.3. Tree G Limita	rowth and Su	rvival: Ke	ey Observa	tions, Ui	ncertaintie	es and
, Q	Looking across th	a array of eve	arimontal	addition st	idios and	tha thraa r	acant
0	observational (or gradien	t) studies we r	ote a num	ber of key	observati	one and as	sociated
9 10	un sorteinties and limitati	angi		iber of key	UUSCI Vali	ons and as	sociated
10	uncertainties and limitati	ons:					
11	Ex	perimental Ad	dition Stu	dies of Tre	e Growth	/Survival	
12 13	• Some studies which can inf	additionally re orm interpretat	ported soi ion of resj	l chemistry ponses that	and/or tr may rela	ee cellular te to geolo	responses, gy and soil
14	chemistry in t	hose locations.					
15 16 17	• <u>S or S + N ad</u> with a small s yellow poplar	<u>dition:</u> Some m et of eastern sp , black cherry,	nultiyear S becies, inc have not i	or S+N ad luding suga reported de	ldition ex ar maple, trimental	periments aspen, whi growth eff	(>20 kg/ha-yr) ite spruce, fect (Table 5B-1
18 19 20	Bethers et al 2 2014). Some [Bethers et al.	2009; Moore ar reported increa , 2009]), while	nd Houle 2 sed growt one report	2013; Jung h (25.2 kgľ rted reduce	and Char N + 28.8 I d growth	ig, 2012; Jo kg S/ha-yr in three sp	ensen et al for 10 years Jecies after 10
21	years that reso	olved in two of	the specie	es after 22	years (Jer	isen et al.,	2014).
22 23 24	• <u>N addition:</u> Se eastern specie 2004; McNult	everal studies f es including oal ty et al 2004; P	ound varia (s, spruce, regitzer et	able results maples an al 2008; a	for grow d pines ( nd Walla	th and surv Fable 5B-1 ce et al 200	vival for several ; Magill et al )7).

1	Observational/Gradient Studies of Tree Growth/ Survival
2 3	Newly available in this review are three large observational studies of tree growth/survival and S/N deposition.
4 • 5 6 7	Although ozone was analyzed in one of the three studies, soil characteristics and other factors with potential to impact tree growth and survival (other than climate) were not assessed. The reason for this is that many of these soil factors are not available nationally in the FIA database.
8 • 9 10 11 12 13 14	<u>S deposition:</u> Two large studies that analyzed growth and/or survival measurements in 94 and 267 species, respectively, at sites across the country, or in the eastern half of the country, describe negative associations of tree survival and growth with the S deposition metric for nearly half the species individually and negative associations of tree survival for 9 of the 10 species' functional type groupings (Dietze and Moorcroft, 2011; Horn et al., 2018). Survival for the same 9 species groups was also negatively associated with long-term average ozone (Dietze and Moorcroft, 2011).
15 16 17 18 19 20	<ul> <li>The S deposition metrics were derived from estimates for total S or sulfate in overlapping time periods of roughly 10 years and include areas, particularly in the eastern U.S., that have experienced decades of much higher deposition.</li> <li>The full range of average SO<sub>4</sub> deposition estimated for the 1994-2005 time period assessed by Dietze and Moorcroft (2011) for the eastern U.S. study area was 4 to 30 kg S ha<sup>-1</sup>yr<sup>-1</sup>.</li> </ul>
21 22 23 24	<ul> <li>Median average S deposition (2000-13) estimated at sites of nonwestern species with neg associations with growth or survival ranged from 5 to 12 kg S ha<sup>-1</sup>yr<sup>-1</sup>, with few exceptions (Horn et al., 2018).</li> </ul>
25 26 27 28 29	<ul> <li>The extent to which the differences in growth or survival across sites with different deposition estimates relate to historically higher deposition at the sites (e.g., <i>versus</i> the deposition metrics analyzed) is unknown. There are few available studies describing recovery of historically impacted sites (e.g., ISA, section IS.4.1, IS.5.1, IS.11.2).</li> </ul>
30 • 31 32 33	<u>N deposition:</u> Three large studies that analyzed growth and/or survival measurements in 24 to 267 species at sites in the northeastern or eastern U.S., or across the country, describe associations of tree survival and growth with several N deposition metrics (Dietze and Moorcroft, 2011; Thomas et al., 2010; Horn et al., 2018).
34 35 36	<ul> <li>The N deposition metrics were derived from estimates for total N or nitrate in overlapping time periods and include areas that have experienced decades of much higher deposition.</li> </ul>
37 38 39	<ul> <li>The full range of average NO<sub>3</sub> deposition estimated for the 1994-2005 time period assessed by Dietze and Moorcroft (2011) for the eastern U.S. study area was 6 to 16 kg N ha<sup>-1</sup>yr<sup>-1</sup>.</li> </ul>
40 41 42 43	<ul> <li>Median average N deposition (2000-13) estimated at sites of nonwestern species for which associations with growth or survival were negative (either over full range or at median for species) ranged from 7 to 12 kg N ha<sup>-1</sup>yr<sup>-1</sup> (Horn et al., 2018).</li> </ul>

1 2 3 4 5 6 7 8	<ul> <li>Median average N deposition (2000-13) estimated at sites of nonwestern species for which associations with growth or survival were positive (either over full range or at median for species) ranged from 7 to 12 kg N ha<sup>-1</sup>yr<sup>-1</sup> (Horn et al., 2018).</li> <li>The extent to which the associations of growth or survival with site-specific N deposition estimates relate to historic patterns of N or S deposition at the sites (e.g., <i>versus</i> the specific magnitude of the N deposition metrics analyzed) is unknown.</li> </ul>					
9	Only a very small subset of the 71 species of Horn et al $(2018)$ have been previously					
10	studied with regard to S deposition and growth or survival, although the study by Dietze and					
11	Moorcroft (2011) included these species in its groupings by plant functional type (Table 5B-6).					
12	With regard to relationships of tree growth or survival with N deposition metrics, some of the					
13	Horn et al (2018) species were also assessed in the study by Thomas et al (2010), as well as all of					
14	the species being included in the groupings of Dietze and Moorcroft (2011). Table 5B-6					
15	indicates a similarity in the findings, particularly of Horn et al (2018) and Dietze and Moorcroft					
16	(2011), although the time period and estimation approach for S and N deposition differ.					
17	Given the role of deposition in causing soil conditions that affect tree growth and					
18	survival, and a general similarity of spatial variation of recent deposition to historic deposition,					
19	the similarity in the two studies' finding may indicate the two different metrics to both be					
20	reflecting geographic variation in impacts stemming from historic deposition. Although the					
21	spatial patterns are somewhat similar, the magnitudes of S and N deposition in the U.S. has					
22	changed appreciably over the time period covered by these studies. An example of this is					
23	illustrated by the patterns of wet deposition of SO4 and NO3 in Figures 5B-9 and 5B-10,					
24	respectively, and patterns of total S and N deposition in Figures 5B-11 and 5B-12. The					
25	appreciable differences in magnitude across the time periods contribute uncertainty to					
26	interpretations related to specific magnitudes of deposition associated with patterns of tree					
27	growth and survival.					
28	Differences in findings of Thomas et al. (2010) may be related to the much shorter N					
29	deposition time period used, as compared to those of Horn et al. (2018) and Dietze and					
30	Moorcroft (2011). The findings unimodal or hump-shape associations for Horn et al. (2018) for					
31	species with positive or negative associations in Thomas et al. (2010) may also reflect different					
32	time periods assessed. Positive effects in Thomas could translate to negative effects in Horn if					
33	the N deposition effects accumulate through time, as hypothesized in Aber et al. 1998. Further,					
34	the occurrence of negative and positive survival or growth associations from Thomas et al (2010)					
35	and Horn et al. (2018) for species in a plant functional grouping for which Dietze and Moorcroft					
36	(2011) found negative association may reflect difference in study areas, e.g., early successional					
37	hardwood, which had positive association of survival with N, includes quaking aspen for which					

- 1 Thomas et al. (2010) reported negative survival association. The study area of Thomas et al.
- 2 (2010) was limited to the Northeast, however, while aspen is prevalent in the Northern Forests
- 3 ecoregion, which is included in Dietze and Moorcroft (2011) study area.

	S Dep	osition		N Deposition	
Species	Dietz and Moorcroft (2011) (SO <sub>4</sub> , wet, 1994-2005)	Horn et al. (2018) (total S, ~2000- 2013)	Dietze and Moorcroft (2011) (NO <sub>3</sub> , wet, 1994-2005)	Thomas et al. (2010) (total N, 2000-2004, FIA data,1970s-90s)	Horn et al. (2018) (total N, ~2000- 2013)
	Positive (↑	) or negative	(↓)association	for growth (G) or surv	vival <sup>a</sup> (Su)
Early Successional Hardwood	↓Su		∱Su		
Betula alleghaniensis, yellow birch		↓Su		Small ↓Su	↓G
Betula lenta		↓Su ↓G			U Su
Betula papyrifera, paper birch		↓Su ↓G		Small ↑Su	U Su
Gleditsia triacanthos		↓G			
Liquidambar styraciflua		↓Su			U Su
Maclura pomifera		↓G			
Populus grandidentata, bigtooth aspen		↓Su ↓G		Small ↓Su	U Su
Populus tremuloides, quaking aspen		↓Su		↓Su ↑G	U Su U G
Prunus serotina, black cherry				∱Su ∱G	U Su U G
Salix nigra		↓G			UG
Late Successional Hardwood	↓Su		∱Su		
Acer negundo, boxelder		↓Su ↓G			
Acer rubrum, red maple		↓Su ↓G		small ↑Su ↑G	↑G
Acer saccharum, sugar maple		↓Su		↑G	
Acer saccharinum, silver maple		↓G			↑G
Carpinus caroliniana, American hornbeam		↓Su ↓G			
Oxydendrum arboreum, sourwood		↓Su			U Su
Tilia americana, American basswood		↓Su ↓G		Small ↓Su	∱G
Northern Midsuccessional Hardwood			↓Su		
Celtis occidentalis, hackberry		↓Su			UG
Fraxinus americana, white ash				↑G	U Su ↑G
Fraxinus pennsylvanica, green ash		.∐Su ∬G			U Su U G
Juglans nigra, black walnut		JG			U Su
Quercus alba, white oak		↓Su			U Su
Quercus ellipsoidalis, northern pin oak		↓G			↑G
Quercus rubra, northern red oak		· · · ·		small ↓Su ↑G	U Su ↑G
Quercus velutina, black oak		↓Su			↑G
Sassafras albidum, sassafras		↓Su			↑G
Ulmus americana, American elm		↓Su↓G			↓Su ↑G
Ulmus rubra, slippery elm		↓Su ↓G			
Hydric	↓Su		∱Su		
Nyssa aquatica		↓G			
Nyssa biflora		↓Su			UG
Taxodium ascendens					↑G
Taxodium distichum		.∐Su ∐G			

# 1 Table 5B-6. Significant associations in the three studies using USFS tree measurements.

	S Dep	osition		N Deposition	
Species	Dietz and Moorcroft (2011) (SO <sub>4</sub> , wet, 1994-2005)	Horn et al. (2018) (total S, ~2000- 2013)	Dietze and Moorcroft (2011) (NO <sub>3</sub> , wet, 1994-2005)	Thomas et al. (2010) (total N, 2000-2004, FIA data,1970s-90s)	Horn et al. (2018) (total N, ~2000- 2013)
	Positive (↑	) or negative	(↓)association	for growth (G) or surv	′ival^(Su)
Southern Midsuccessional Hardwood	↓Su		↑Su		
Carya alba, mockernut hickory					↓Su
Carya glabra, pignut hickory		↓Su		∱G	
Carya texana, black hickory					↑G
Liriodendron tulipifera, yellow poplar		↓Su		∱G	↑G
Nyssa sylvatica, black gum				·	U Su
Quercus coccinea, scarlet oak				↓Su ↑G	U Su U G
Quercus falcata, southern red oak					↓Su
Quercus laurifolia, laurel oak		↓Su			
Quercus muelenbergii, chinkapin oak					U Su
Quercus nigra, water oak		↓Su ↓G			↓Su U G
Quercus prinus, chestnut oak		↓Su		Small ↓Su	UG
Quercus stellata, post oak					U Su
Ulmus alata, winged elm		↓Su			
Evergreen Hardwood	↓Su		∱Su		
Magnolia virginia		↓Su			UG
Midsuccessional Conifer			∱Su		
Picea rubens, red spruce				Small ↓G	
Picea glauca, white spruce		↓G			
Pseudotsuga menziesii, Douglas fir		↓G			U Su ↑G
Late Successional Conifer	Weak ↑ Su		∱Su		
Abies balsamea, balsam fir				↑G	
Juniperus virginiana, eastern redcedar		↓Su ↓G			U Su
Thuja occidentalis, northern white cedar				Small <sup>в</sup> ↓G	
Tsuga canadensis, eastern hemlock		↓G			
Northern Pine	↓Su		∱Su		
Pinus resinosa, red pine		↓Su ↓G		↓G	U Su, ↑G
Pinus regida, pitch pine		↓Su			UG
Pinus strobus, eastern white pine		↓Su ↓G		small ↓Su small ↑G	↑G
Southern Pine	↓Su		∱Su		
Pinus echinata, shortleaf pine		↓Su ↓G			
Pinus elliotti, slash pine		↓Su ↓G			US
Pinus palustris, longleaf pine		↓Su			UG
Pinus taeda, loblolly pine		↓Su ↓G			
Pinus virginiana, Virginia pine		↓Su		<u> </u>	U Su
A For Dietze and Moorcroft (2011), an up arrow is shown for survival if they reported a negative association with mortality. B For Thomas et al. (2010), "small" used when growth or survival response per unit N is <1%e.					

For Horn et al. (2018) "U" used for unimodal (or hump-shaped) associations (positive at lower deposition values and negative at higher).



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Figure 5B-9. Annual mean wet SO<sub>4</sub> deposition in the U.S. for 1989-1991 (top panel) and 2014-2016 (bottom panel) (U.S. EPA, 2023; NADP, 2018).



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6

2014-2016 (bottom panel) (U.S. EPA, 2023; NADP, 2018).



# Figure 5B-11. Wet plus dry deposition of total sulfur over 3-year periods. Top: 2000-2002; Bottom: 2016-2018. Drawn from the ISA, Figure 2-70.





# **5B.3 SPECIES RICHNESS OF HERB AND SHRUB COMMUNITIES**

The subsections below summarize salient aspects of studies that have assessed herb and shrub community metrics and their relationship to N deposition. The addition studies in section 5B.3.1 below evaluated the impact of fertilizer treatments using ammonium nitrate. Section 5B.3.2 summarizes the few recent observational studies that statistically analyze variation in species richness metrics with variation in N deposition, while also providing detailed information regarding the largest such study (Simkin et al., 2016). The extent to which the observational studies account for potential influence of S deposition varies.

9

## 5B.3.1. Experimental Addition Studies

- 10 A number of experimental addition studies focused on N (e.g., through addition of
- 11 ammonium nitrate fertilizer) are discussed in the ISA and summarized in Table 5B-7 below.

Location	Description	Additions	Findings
Joshua Tree National Park, in Mojave desert, CA (Allen et al., 2009)	Assessed biomass and % cover responses of native and non-native grasses to two fertilization levels at four sites	5 and 30 kg N ha <sup>-1</sup> yr <sup>-1</sup> as ammonium nitrate (NH <sub>4</sub> NO <sub>3</sub> ) fertilizer over 2 years Ambient deposition was estimated to be approximately 5 – 8 kg N ha <sup>-1</sup> yr <sup>-1</sup>	In 1st year, non-native grass biomass increased significantly at three of the four study sites receiving 30 kg N/ha/yr. No significant change with 5 kg N/ha/yr; of with either dose in 2 <sup>nd</sup> year. No change in % cover. Native grass species richness increased with 30 kg N/ha-yr at 1 site that authors judged related to lower nonnative species presence.
Prairie grasslands in Cedar Creek Ecosystem Science Reserve, MN Clark and Tillman (2008)	Study plots in two prairie- like successional grasslands and one native savanna grassland. The soils were limed to maintain constant pH (and avoid acidification).	10, 20, 34, 54 and 95 kg N ha <sup>-1</sup> yr <sup>-1</sup> (ammonium nitrate addition) over 23 years (1982 to 2004). Background wet deposition of N was estimated to have averaged 6 kg N ha <sup>-1</sup> yr <sup>-1</sup> wet deposition.	Species numbers declined with increasing chronic addition, including at the lowest addition (10 kg N/ha/yr). In a subset of plots for which additions were ceased after 10 years, relative species numbers increased, converging with controls after 13 years due to losses of species in controls.
Dry sedge meadow in Rocky Mountain National Park, CO Bowman et al (2012)	Five replicate plots (20 total) in a dry meadow community. Study assessed plant species richness, cover of vascular plants, above ground biomass, and soil chemistry.	5, 10 and 30 kg N ha <sup>-1</sup> yr <sup>-1</sup> (ammonium nitrate addition) over 4 years starting in 2006. Background deposition was estimated to be 4 kg N ha <sup>-1</sup> yr <sup>-1</sup>	No significant effect on plant species richness or diversity. No significant effect on foliar % N or above ground biomass. Based on <i>Carex rupestris</i> increasing in cover from 34 to 125% in response to additions, authors estimated 3 kg N/ha-yr as deposition associated with an increase in <i>C rupestris</i> cover and 9 - 14 kg N/ha-yr with NO <sub>3</sub> -leaching in soil solution.

#### 12 Table 5B-7. Experimental addition studies assessing herb and shrub community responses.

Location	Description	Additions	Findings			
Santa Margarita Ecological Reserve, Riverside, California Vourlitis, 2017	Study of long term effects of N deposition on native and exotic plant cover in coastal sage scrub communities. 4 control and 4 addition plots (10 x 10 m)	50 kg N ha <sup>-1</sup> yr <sup>-1</sup> over 13 years. Background deposition estimated at 4 – 6 kg N ha <sup>-1</sup> yr <sup>-1</sup>	Increase in the native shrub Artemesia californica in the 4th and 5–9th yr of the 13-yr experiment; decrease in the native shrub Salvia mellifera in the 4th and 11–13th yr; increase in the exotic plant Brassica nigra in the 11–13 <sup>th</sup> yr			
Santa Margarita Ecological Reserve, Riverside, California and Sky Oaks Field Station, San Diego County, CA Vourlitis and Pasquini, 2009	Study of effects of N deposition on plant community composition in coastal sage scrub and chaparral communities. 4 control and 4 addition plots (10 x 10 m) at each site (16 total)	50 kg N ha <sup>-1</sup> yr <sup>-1</sup> for 5 years as granular NH <sub>4</sub> NO <sub>3</sub> (2003–2006) or (NH <sub>4</sub> )2SO <sub>4</sub> (2007–2008). Background deposition estimated as 6-8 kg N ha <sup>-1</sup> yr <sup>-1</sup>	Dry season addition of N significantly changed community composition in coastal sage scrub communities, but not in chaparral communities			
Great Basin, California Concilio and Loik 2013	Study effects of elevated N deposition on sagebrush steppe communities in 54 paired plots (half control, half with additions).	50 kg N ha <sup>-1</sup> yr <sup>-1</sup> for 4 years starting in 2007. Background deposition estimated as 1 – 3 kg N ha <sup>-1</sup> yr <sup>-1</sup>	Community composition (native species diversity and abundance of the invasive grass <i>Bromus tectorum</i> ) differed by disturbance history (e.g. fire), but was not affected by N deposition.			
Sevilleta National Wildlife Refuge, New Mexico Collins et al., 2017	Study of the effect of nighttime warming, winter precipitation and N deposition in 40 plots (3.0 x 3.5 m each) randomly crossed across treatment effect.	20 kg N ha <sup>-1</sup> yr <sup>-1</sup> for 7 years starting in 2006. A wildfire burned the plots after the second year. Ambient deposition was approximately 3 kg N ha <sup>-1</sup> yr <sup>-1</sup> A	Native desert grass communities were affected by N deposition in the 3 years following the fire, but not in the two years preceding the fire or the last year of the experiment.			
Arches National Park, Colorado Plateau, Utah McHugh et al 2017	Study of community composition in a semi- arid grassland d deposition was not reported in	0, 2, 5 and 8 kg N ha <sup>-1</sup> yr <sup>-1</sup> for 2 years starting in 2011. Background deposition was estimated as 2 – 3 kg N ha <sup>-1</sup> yr <sup>-1</sup> n this publication, we have estim	No significant change in community composition or species richness, but did find a strong connection between composition and soil microbial community structure.			
based on TDEP version 2018.02, using EPA's CL Mapper Tool at: https://www.epa.gov/gcx/about-cl-mapper.						

## **5B.3.2.** Gradient or Observational Studies

Recent gradient studies have included analyses investigating the potential of N

3 enrichment in southern California to alter plant community composition through increases in the

4 presence of invasive annual species (ISA, Appendix 6, section 6.3.6). A recent study by Cox et

- 1 al. (2014) utilized a landscape-level analysis of vegetation change since the 1930s to investigate
- 2 risk of conversion of coastal sage scrub vegetation to exotic annual grassland and any association
- 3 with N deposition. The authors concluded that sites with 2002 N deposition estimates (based on
- 4 CMAQ modeling [Tonnesen et al., 2007]) less than 11 kg N ha<sup>-1</sup>yr<sup>-1</sup> were less likely to have
- 5 converted from Coastal sage scrub to non-native grasslands (ISA, Appendix 6, section 6.3.6; Cox
- 6 et al., 2014). The authors also evaluated the circumstances associated with recovery of coastal
- 7 sage scrub communities from exotic annual grassland that was observed in the 1930s maps, and
- 8 reported that plots in areas where surrounding plots had little or no exotic grassland and 60%
- 9 cover by coastal sage scrub had increased probability of recovery (Cox et al., 2014). A second
- 10 study across the same gradient of 2002 N deposition estimates (6.6 to 20.2 kg N ha<sup>-1</sup>yr<sup>-1</sup>)
- 11 reported similar observations, finding that sites with N deposition above 10 kg N/ha-yr had lower
- 12 native species richness (Fenn et al., 2010).
- 13 One of the largest studies, by Simkin et al. (2016), analyzed relationships between

14 observed variation in herb and shrub species richness and average N deposition, soil pH, and

15 annual average temperature and precipitation at more than 15,000 forest, woodland, shrubland

16 and grassland sites in multiple regions of the U.S. (Figure 5B-13; Table 5B-8). The study

- 17 categorized sites into open-canopy and closed-canopy communities and, in a "national" analysis,
- 18 investigated quantitative relationships between site variation in species richness, assessed over
- 19 the 23-year period from 1990 to 2013, and in estimates of average N deposition for the "modern"
- 20 period of 1985 to 2011 (Simkin et al., 2016, Supplemental Information, SI Methods).

## 21 Table 5B-8. Key aspects of analysis by Simkin et al. (2016)

Study Area	Community	N Deposition estimates	Other variables considered
	assessments		
Northwestern U.S. (predominantly	Assessments	10-yr average (2002-11)	Soil pH, precipitation and
WA, OR, far north CA, western		dry deposition from	temperature (1981-2010)
MT, NV, UT), northeastern CO,	>15,000 sites	CMAQ added to 27-year	
MI, mid-Atlantic (MD, VA) and		average (1985-2011) wet	
Southeast (NC, SC, GA, FL)		deposition from NADP.	

The site assessments were drawn from seven databases of biological survey data sources, with varied distribution across the states represented. For example, more than a third of the sites were in Minnesota and the Pacific Northwest (WA and OR) and another third in the Carolinas and Virginia; about 100 sites are in the northeastern U.S. (Simkin et al., 2016, Supplemental

26 Information, Table S1; Figure 5B-13).



#### Figure 5B-13. Sites included in analysis by Simkin et al. (2016). Based on dataset available at https://datadryad.org/stash/dataset/doi:10.5061/dryad.7kn53

4 When sites were grouped as closed-canopy (forested) sites vs open-canopy (woodland, 5 shrubland and grassland) sites, a statistical relationship was observed for variation in herbaceous 6 species richness (number of herbaceous species) with variation in N deposition (and soil pH, 7 followed by temperature and precipitation). Different quantitative relationships were observed 8 for the two categories of sites. In open-canopy ecosystems, there was a positive relationship 9 between herbaceous species richness and N deposition at the low end of the deposition range 10 (sites with higher N deposition had more species), then a negative relationship with N deposition values above an average of 8.7 kg N ha<sup>-1</sup>yr<sup>-1</sup> (Simkin et al., 2016). In closed-canopy ecosystems, 11 12 the variation in forest understory species richness with variation in N deposition was highly 13 dependent on soil pH. At sites with low pH (4.5) and N deposition above 11.6 kg N ha<sup>-1</sup>yr<sup>-1</sup>, a 14 negative relationship was observed for species richness with N deposition (higher N deposition 15 sites had lower species richness). At sites with basic soils (pH > 8.0), no negative association of 16 species richness with N deposition was observed across the full range of N deposition estimates, which extended up to about 20 kg N/ha-yr (Simkin et al., 2016). 17 18 The statistical models for the two categories of sites were then applied to the pH,

19 temperature and precipitation for each site to predict N deposition values expected to be

- 1 associated with the point at which species losses begin given the site conditions.. For the forested (closed-canopy sites), these N deposition values ranged from 7.9 to 19.6 kg N ha<sup>-1</sup>yr<sup>-1</sup>, with a 2 3 mean of 13.4 kg N ha<sup>-1</sup>yr<sup>-1</sup>. Across the open-canopy sites, these N deposition values ranged from 4 7.4 to 10.3 kg N ha<sup>-1</sup>yr<sup>-1</sup>, with a mean of 8.7 kg N ha<sup>-1</sup>yr<sup>-1</sup> (Simkin et al., 2016). 5 Simkin et al. (2016) also performed regional gradient analyses for a set of sites for which 6 the data were judged sufficient. This involved 44 gradients for a subset of 26 vegetation types 7 that spanned a range in N deposition estimates the authors judged to be adequate. Of the 44 8 gradients, a negative association of species richness with N dep was observed at 16 (36.5%), a 9 positive association at 8 (18%), and no association found for the remaining 20 (45%). Among the 10 8 gradients showing positive associations, most had N deposition estimates averaging at or below 3 kg N ha<sup>-1</sup>vr<sup>-1</sup>. Overall, a negative association of species richness w N deposition estimates was 11 12 more common for gradients involving soil that was acidic, higher precipitation or warmer 13 temperatures (Simkin et al., 2016). 14 In summary, the national-scale analysis of herbaceous species richness by Simkin et al. 15 (2016) indicated that N deposition effects on forest closed-canopy species richness is highly 16 dependent on soil pH (ISA, Appendix 6, section 6.3.3.2). At open-canopy sites (e.g., grasslands, shrublands, and woodlands) with low rates of N deposition (e.g., below 6.5 kg N ha<sup>-1</sup>yr<sup>-1</sup> for soil 17 pH of 4.5 and below 8.8 kg N ha<sup>-1</sup>yr<sup>-1</sup> for soil pH of 7), relatively higher N deposition was 18 19 generally associated with higher plant species richness (Simkin et al., 2016; ISA, Appendix 6, 20 section 6.3.5). With N deposition above 8.7 kg N ha<sup>-1</sup> yr<sup>-1</sup>, on average across site conditions, there were lower levels of species richness. At forested sites, relatively higher N deposition was 21 22 associated with higher plant species richness for sites with soil pH of 4.5 and N deposition 23 estimates below 11.6 kg N ha<sup>-1</sup>yr<sup>-1</sup>. With N deposition above this level there was a reduction in 24 species richness. At forested sites with basic soil, no value of N deposition was negatively associated with species richness up to 20 kg N ha<sup>-1</sup> yr<sup>-1</sup>. At both the national and gradient 25 analyses, sites with N deposition estimates at or below 3 kg N ha<sup>-1</sup>yr<sup>-1</sup> showed little or no 26 27 reduction in species richness with N deposition (Simkin et al., 2016). It is important to note that 28 species richness is merely the count of the number of species at a site. The national results show 29 that at lower N deposition levels, there are more species gained than lost as N deposition increases up to point (e.g., 8.7 kg N ha<sup>-1</sup> yr<sup>-1</sup> for open canopy on average), above that level there 30 are more species lost than gained. Thus, there could be species lost at all levels of N deposition 31 32 (Clark et al. (2008), only a species-level analysis would show whether there were individual 33 species lost at these lower levels that may have been masked in the total count of species. 34 Study limitations with regard to interpretations specific for N deposition include that no 35 other pollutants with potential to affect species richness (and which may covary in many places 36 with N deposition), including sulfate and ozone, were considered. Further, the "modern" N
  - May 2023

1 deposition estimates (1985-2011) were correlated with both shorter duration more recent

- 2 estimates and with longer duration historical estimates, introducing uncertainty with regard to the
- 3 particular deposition of interest with greatest influence on the results. This correlation coupled
- 4 with the variation in magnitude of the deposition estimates for the various periods also
- 5 contributes uncertainty regarding identification of what might be termed N deposition thresholds
- 6 that might contribute to different types of relationships with species richness. Further, the study
- 7 does not provide information on the species that are absent vs present, or their role in the
- 8 community, across the varying species richness values. Additionally, site distribution was
- 9 heterogeneous across parts of the U.S. For example, the most densely sampled closed canopy
- 10 areas were in the southern Appalachians and Virginia, and Minnesota, areas of historically high
- 11 and low deposition, respectively (Figure 5B-13). With regard to herb an shrub communities,
- 12 there was appreciable representation in Minnesota and virtually no representation in
- 13 Mediterranean California or the Great Plains. The potential influence of the relative distribution
- 14 of sites across areas of greater versus lesser historical deposition is unclear.

# 15 5B.4 LICHEN COMMUNITY COMPOSITION

Lichens absorb N, S, and other elements from the air and from material deposited on their surfaces. Accordingly, lichens can be sensitive to air pollution and are frequently used as indicators of air quality, and associated deposition (2008 ISA, section 3.3.5.1), on forest ecosystems. Shifts in lichen community composition to greater presence of more N tolerant species have been associated with areas that have received high acidifying deposition and high concentrations of SO<sub>2</sub>, N oxides and reduced N, such as the eastern U.S. (2008 ISA, section 3.2.2.3).

23 Research in the late 1970s-early 1980s reported inverse associations of lichen cover with 24 atmospheric oxidants in the San Bernardino Mountains just outside Los Angeles, California. 25 Studies in this region have reported a reduction in lichen species by about 50% since the early 26 1900s, with elevated HNO<sub>3</sub><sup>-</sup> identified as a contributor to lichen community declines in the Los 27 Angeles basin dating back to the 1970s. Studies since the 2008 ISA indicate these communities 28 have not yet recovered (ISA, Appendix 3, section 3.3). Surveys of urban and industrial areas in 29 the 1970s and 80s (e.g., in urban areas of Great Britain) also identified SO<sub>2</sub> as a factor in lichen 30 community declines observed lichen deaths (ISA, Appendix 3, section 3.2; Hutchinson et al., 31 1996<sup>16</sup>;). The relative influences of airborne versus deposited air pollutants in such impacts is 32 unclear.

<sup>&</sup>lt;sup>16</sup> The publication by Hutchinson et al., 1996, cited in ISA, cites to Seaward (1987) as the support for its characterization; the characterization summarized here is also drawing on the specific details provided by Seaward (1987).

#### 5B.4.1. Studies Investigating Direct Effects of Pollutants in Ambient Air

2 Sulfur oxides and oxides of N have been associated with effects on lichens (ISA, 3 Appendix 3, section 3.2 and 3.3). In laboratory experiments involving daily HNO<sub>3</sub> exposures, 4 with peaks near 50 ppb, over durations of 18 to 78 days, effects on lichen photosynthesis were 5 reported, among other effects (ISA, Appendix 6, section 6.2.3.3; Riddell et al., 2012). Based on 6 studies extending back to the 1980s, HNO<sub>3</sub> has been suspected to have had an important role in 7 the dramatic declines of lichen communities that occurred in the Los Angeles basin (ISA, 8 Appendix 3, section 3.4; Nash and Sigal, 1999; Riddell et al., 2008; Riddell et al., 2012). For 9 example lichen transplanted from clean air habitats to analogous habitats in the Los Angeles 10 basin in 1985-86 were affected in a few weeks by mortality and appreciable accumulation of H+ 11 and NO<sub>3</sub><sup>-</sup>(ISA, Appendix 3, section 3.4; Boonpragob et al., 1989). 12 Air monitoring data summarized in Chapter 2 indicate areas of the U.S. experiencing 13 appreciably higher annual mean NO<sub>2</sub> concentrations in the 1980s compared to more recent years (Figure 2-21). For example the 95<sup>th</sup> percentile of U.S. sites ranged from just over 50 ppb to just 14 15 over 60 ppb during the 1980s (Figure 2-21). During the 1980s, and earlier, the Los Angeles 16 metropolitan statistical area (MSA) had some of the highest annual average NO<sub>2</sub> concentrations. 17 For example, the annual average NO<sub>2</sub> concentration in Los Angeles was 0.078 ppm in 1979, 18 0.071 ppm in 1980, 0.058 ppm in 1985 and 0.057 ppm in 1989 (U.S. EPA, 1983, 1987, 1991). 19 Concentrations of  $O_3$  in Los Angeles were also quite high during this time (U.S. EPA, 1983, 20 1987, 1991), however, while  $O_3$  impacts on plants is well established, research with lichens 21 indicates a lesser sensitivity. This contributes to the evidence for NO<sub>2</sub>, and particularly, HNO<sub>3</sub>, as 22 "the main agent of decline of lichen in the Los Angeles basin" (ISA, Appendix 3, p. 3-15). 23 Co-occurring elevations in  $SO_2$  and ozone contribute uncertainty to identification of a

threshold concentration of N oxides likely to elicit lichen community changes such as those that occurred in the Los Angeles basin. More recent studies indicate variation in eutrophic lichen abundance to be associated with variation in N deposition metrics (ISA, Appendix 6, section 6.2.3.3). The extent to which these associations are influenced by residual impacts of historic air quality is unclear.

29 30

# 5B.4.2. Observational Studies Investigating Relationships with Atmospheric Deposition

Several recent studies have reported negative associations of lichen community
composition/abundance and N deposition (and S deposition) metric values in areas of the
Northwest, California and at some sites in the northeast (Table 5B-9; ISA, Appendix 6, section
6.5). For example, analyses of surveys in 1990s report species richness differences among sites
in Pacific NW to vary with estimates of N deposition (and N-PM<sub>2.5</sub>) across sample sites ranging

1 from approximately 8.2 to <1 kg N ha<sup>-1</sup>yr<sup>-1</sup> and 10 to <1 kg dissolved inorganic N ha<sup>-1</sup>yr<sup>-1</sup>

- 2 (Geiser et al., 2010; Root et al., 2015, Appendix B, Table B.1). The study by Geiser et al. (2010)
- 3 analyzed relationships between lichen community composition and several N deposition metrics
- 4 at sites in Western Oregon and Washington forests. At other sites in the western U.S., Root et al.
- 5 (2015) analyzed relationships between lichen community/abundance metrics and lichen N
- 6 concentrations and N deposition estimates extrapolated from lichen N concentrations. Statistical
- 7 modeling was used to identify N deposition estimates associated with a change in lichen
- 8 community/abundance metric(s) for sites in 2 ecoregions. Both papers utilized a linear regression
- 9 approach. Geiser et al. (2010) used the regression to relate community composition to an "air
- 10 score," while Root et al. (2015) used it to relate a community-based index to air concentrations
- 11 of nitrogen in fine PM, which was then related to N deposition.

12 There are several limitations associated with use of these studies' findings for purposes of 13 interpreting potential risk to lichens of recent N deposition. For example, the estimates of 14 deposition utilized different methods than the current commonly accepted methods. The potential 15 role of other unaccounted environmental factors (including ozone, SO<sub>2</sub> and S deposition) has not 16 been addressed in these observational/gradient, uncontrolled studies, and there is a scarcity of 17 controlled N addition experiments that might augment conclusions The significance of findings 18 of the western studies is unclear for other areas of the U.S., and there is uncertainty concerning 19 the independence of any effect of the deposition levels analyzed from residual effects of past N 20 deposition. Further, the extent to which these observations reflect communities still exhibiting 21 impacts of much higher pollution of the 1970s-80s is unknown. Although some studies have 22 investigated historical impacts, there remain uncertainties as to the extent to which impacts on 23 lichen communities noted in recent studies reflect recent N deposition. And there are few 24 controlled N addition experiments that might augment or inform interpretation of the findings of 25 observational/gradient studies. Other studies in Europe and Canada have not reported such 26 associations with relatively large N deposition gradients.

# 1 Table 5B-9. Lichen endpoints and associated deposition estimates.

Description	Deposition	Findings
	Estimates	
Cleavitt et al. (2011) analyzed 4 plots distributed across a gradient in estimated S deposition in Acadia National Park, ME	12 to 18 kg S/ha-yr	Rather than relate deposition to lichen distribution, this study reported that throughfall chemistry influenced bark pH and that influenced the suitability of tree boles as habitat for lichen. Epiphytic lichen species richness and presence of pollution-sensitive epiphytes were greater on red maple trees, which have a higher pH in the bark relative to red spruce trees.
Cleavitt et al (2015) analyzed 24 sites in 4 Class I areas in Northeastern U.S. (Lye Brook Wilderness, VT, Great Gulf and Presidential Range-Dry River Wiildernesses, NH, and Acadia National Park, ME); assessed multiple metrics for lichen status associations with concurrent (2-yr ave) and cumulative (2000-13) S and N deposition estimates. Cumulative and 2-yr average recent N deposition were tightly correlated (r2=0.90 p , 0.0001);cumulative and recent S deposition were not correlated. Aerosol NO <sub>3</sub> <sup>-</sup> declined from ~0.7-0.9 to ~0.25- 0.5 ug/m <sup>3</sup> across 14-yr period. Geiser et al (2010) analyzed data at sites in Western OR and WA forests, calculating different N metrics (total, dry and wet N deposition; wet NO <sub>3</sub> +NH <sub>4</sub> deposition for specified breakpoint in "air scores." Statistical modeling of FIA plot air scores based on aspects of lichen community composition and lichen N/S concentrations (assessed 1994-2002) for data subset, considering elevation, precip (1961-90), hardwood basal area (Geiser and Neitlich, 2007). Then model used to predict scores for remaining plots. Range of scores divided into six bins from "best" (lowest bin) to "worst" (highest bin).	Total S deposition of ~6-15 kg S/ha-yr across the 4 areas in 2000; with subsequent reductions to ~3-6 kg S/ha-yr by 2013. Total N deposition of ~4-15 kg N/ha-yr across 4 areas In 2000; with subsequent reductions to ~3-8 kg N/ha-yr (Cleavitt et al., 2015, Figure 4) Average 1990-99 N deposition estimated from CMAQ modeling (0.8 – 8.2 kg/ha-yr across all sites); NADP wet deposition and IMPROVE particulate N for 1994-2002	Negative associations of lichen species richness, abundance of N-sensitive species, and poorer thallus condition with annual mean and cumulative N deposition. Cumulative dry deposition of S yielded best fit to decreases in thallus condition, poorer community-based S Index values, and absence of many S-sensitive species, indicating stronger role for legacy of historical deposition than recent deposition patterns. "Lichen metrics were generally better correlated with cumulative deposition than annual deposition" "In our study, dry S deposition related more closely to patterns in lichen metrics than total or wet S deposition. Dry deposition of S may be more harmful to lichens, both because it has the potential to become highly concentrated when the thallus is rehydrated, and because it largely originates from SO <sub>2</sub> , which has a long history of toxicity to lichens" For breakpoint between 3 <sup>rd</sup> and 4 <sup>th</sup> air scores,total N deposition ranged from about 3 to 9 kg N/ha-yr The score equal to the breakpoint between the 3 <sup>rd</sup> and 4 <sup>th</sup> bins ("fair" and "degraded") was associated with 33-43% fewer oligotrophic species and 3 to 4 fold more eutrophic species than scores in the "best" bin. Per Geiser & Neitlich 2007 for same areas: "Ozone is potentially adversely affecting Pacific Northwest lichens." "Ambient [air] concentrations of NOx often correlate with SO <sub>2</sub> , making it difficult to separate SO <sub>2</sub> effects on lichen communities from NOx effects."
Root et al. (2015) analyzed data for sites in WA, northern ID, NW MT, OR and far NE CA for relationship between lichen community metrics (assessed 1993-2011) and lichen N concentrations (samples 1993-2001) and N deposition estimated from lichen N. Created lichen index relating lichen N to species frequency (excluding uncommon species and species with "ambiguous relationships").	Inorganic N deposition extrapolated from lichen N concentrations, estimated to range from 0.174 to 9.49 kg N/ha-yr across sampling plots	Based on a judgment that "[I]ichen communities did not appear to be strongly impacted by N concentration below 0.378 ug N/m <sup>3</sup> /year" which was the lowest N-PM <sub>2.5</sub> concentration near "known N pollution sources," and the associated lichen N concentration estimated by linear regression, the throughfall N deposition was estimated to be 2.5 kg Ha-yr. Throughfall N deposition estimated from the lichen index value estimated for the chosen N-PM <sub>2.5</sub> and its estimated relationship with throughfall N, was estimated to be 1.5 kg N/ha-yr.

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- 21

1 Attachments 2

1			
2			

## Attachment 1

## **Species by Plant Functional Group**

## 4 Drawn from Dietze and Moorcroft (2011) "Tree mortality in the eastern and 5 central United States: patterns and drivers"

6

Plant Functional Group	Genus Species	Common Name	Genus Species	Common Name		
	Ailanthus altissima	ailanthus	Populus alba	silver poplar		
	Albizia julibrissin	Mimosa	Populus balsamifera	balsam poplar		
	Alnus	alder	Populus deltoides	eastern cottonwood		
	Betula	Birch	Populus deltoides sub monilifera	plains cottonwood		
	Betula alleghaniensis	yellow birch	Populus grandidentata	bigtooth aspen		
	Betula lenta	sweet birch	Populus tremuloides	quaking aspen		
	Betula nigra	river birch	screwbean mesquite			
Farly Successional	Betula papyrifera	paper birch	paper birch Prunus d			
Hardwood	Betula populifolia	gray birch	Prunus americana	American plum		
- Large positive	Bursera simaruba	gumbo limbo	Prunus aviumPRAV	sweet cherry		
influence of SO <sub>4</sub>	Catalpa	catalpa	Prunus nigra	Canada plum		
deposition on	Catalpa bignoniodes	southern catalpa	Prunus pensylvanica	pin cherry		
mortality	Catalpa speciosa	northern catalpa	Prunus serotina	black cherry		
- negative	Elaeagnus angustifolia	Russian-olive	Prunus virginiana	chokecherry		
deposition on	Ficus aurea	Florida strangler fig	Robinia pseudoacacia	black locust		
mortality	Gleditsia triacanthos	honeylocust	Salix	willow		
,	Gymnocladus dioicus	Kentucky coffeetree	Salix alba	white willow		
	Larix laricina	tamarack	Bebb willow			
	Larix spp	Larch spp	costal plain willow			
	Liquidambar styraciflua	sweetgum	Salix nigra	black willow		
	Maclura pomifera	Osage-orange	Salix sepulcralis	weeping willow		
	Melia azedarach	Chinaberrytree	Sideroxylon lanuginosum ssp. lanuginosum	gum bully		
	Paulownia tomentosa	paulownia	Vernicia fordii	tung-oil-tree		
	Populus	poplar				
	Avicennia germinans	Black-mangrove	Magnolia grandifolia	southern magnolia		
Evergreen Hardwoods	Casuarina lepidophloia	belah	Magnolia virginiana	sweetbay		
- Large positive	Cinnamomum camphora	camphor tree	Melaleuca quinquenervia	melaleuca		
deposition on	Conocarpus erectus	buttonwood mangrove	Persea borbonia	redbay		
mortality	Eucalyptus	eucalyptus	Quercus margarettiae	dwarf live oak		
- negative influence	Eucalyptus grandis	grand eucalyptus	Quercus virginiana	live oak		
of NO <sub>3</sub> deposition	Gordonia lasianthus	loblolly-bay	Rhizophora mangle	American mangrove		
on mortality	llex opaca	American holly	Umbellularia californica	California laurel		
	Laguncularia racemosa	white -mangrove				
Lhudria	Carya aquatica	water hickory	Planera aquatica	water elm		
Tyuric	Citrus	Citrus	Populus heterophylla	swamp cottonwood		

Plant Functional Group		Genus Species	Common Name	Genus Species	Common Name		
-	Large positive	Eugenia rhombea	red stopper	Quercus lyrata	overcup oak		
	influence of SO4	Gleditsia aquatica	waterlocust	Sabal palmetto	cabbage palmetto		
	deposition on	Metopium toxiferum	Florida poisontree	Salix amygdaloides	peachleaf willow		
	nonality	NULL	palm, other	Taxodium ascendens	pondcypress		
-	of NO <sub>3</sub> deposition	Nyssa aquatica	water tupelo	Taxodium distichum	baldcypress		
	on mortality	Nyssa biflora	swamp tupelo	Thrinax morrisii	key thatch palm		
	,	Nyssa ogeche	Ogechee tupelo				
Lat	e Successional			Juniperus virginiana var			
Co	nifer	Abies balsamea	Balsam fir	silicicola	Southern redcedar		
-	negative influence	Chamaecyparis thyoides	Atlantic white-cedar	Thuja occidentalis	northern white-cedar		
	of NO <sub>3</sub> deposition	Juniperus	juniper	Tsuga	hemlock		
	on mortality	Juniperus ashei	Ashe juniper	eastern hemlock			
-	influence of SO4	Juniperus scopulorum	Rocky Mountain juniper	Tsuga caroliniana	Carolina hemlock		
	deposition on mortality	Juniperus virginiana	eastern redcedar				
		Acer	Maple	Carpinus caroliniana	hornbeam		
		Acer barbatum	Florida maple	Castanea dentata	American chestnut		
		Acer leucoderme	chalk maple	Cornus florida	Flowering dogwood		
		Acer negundo	boxelder	Diospyros	persimmon		
		Acer nigrum	black maple	Diospyros virginiana	common persimmon		
Lat	e Successional	Acer pensylvanicum	striped maple	Fagus grandifolia	beech		
Ha	rdwood Large positive	Acer platanoides	Norway maple Halesia		silverbell		
-		Acer rubrum	red maple	Halesia carolina	Carolina silverbell		
	deposition on	Acer saccharinum	silver maple	two-wing silverbel			
	mortality	Acer saccharum	sugar maple	sourwood			
-	negative influence	Acer spicatum	mountain maple	Platanus	sycamore		
	of NO <sub>3</sub> deposition			Sapindus saponaria var			
	on mortality	Aesculus	buckeye	drummondii	western soapberry		
		Aesculus flava	yellow buckeye	Tilia	basswood		
		Aesculus glabra	Ohio buckeye	Tilia americana	american basswood		
		Aesculus glabra var		l Illa americana var	Carolina basswood		
		argula		Tilia amoricana var			
		Alnus alutinosa	Furopean alder	heteronhvlla	American basswood		
Mic	Isuccessional	Ahies	fir snn	Picea glauca	white spruce		
COI	nifer	Abies concolor	white fir	Picea mariana	black spruce		
-	negative influence	Abies fraseri	Fraser fir	Picea nungens	Blue spruce		
	of NO <sub>3</sub> deposition	Picea	spruce	Picea ruhens	red spruce		
	on mortality	Picea ahies	Norway Spruce	Psoudotsuga monziesii	Douglas-fir		
		1 1000 00100		,r ooddolddyd mon2ioon			
		Amelanchier	serviceberry	Morus alba	white mulberry		
		Amelanchier arborea	Downy serviceberry	Morus rubra	red mulberry		

Plant Functional Group	Genus Species	Common Name	Genus Species	Common Name
				eastern
	Carya	hickory	Ostrya virginiana	hophornbeam
	Carya cordiformis	bitternut hickory	Quercus alba	white oak
	Carya ovalis	red hickory	Quercus bicolor	swamp white oak
	Carya ovata	shagbark hickory	Quercus ellipsoidalis	northern pin oak
	Celtis laevigata var			
	reticulata	netleaf hackberry	Quercus Ilicifolia	scrub oak
	Celtis occidentalis	hackberry	Quercus macrocarpa	bur oak
	Cladrastis kentukea	yellowwood	Quercus palustris	pin oak
Northern	Crataegus	hawthorn	Quercus princides	swart chinakapin oak
Midsuccessional	Crataegus crus-galli	cockspur hawthorn	Quercus rubra	northern red oak
Hardwood	Crataegus mollis	downy hawthorn	Quercus velutina	black oak
<ul> <li>positive influence</li> </ul>	Fraxinus americana	white ash	Sassafras albidum	sassafras
of NO₃ deposition on mortality	Fraxinus nigra	black ash	Sorbus americana	American mountain- ash
		ave en esh	Carbus susuraria	European mountain-
	Fraxinus pennsylvanica	green asn	Sorbus aucuparia	asn
	Fraxinus protunda	pumpkin asn	Ulmus	eim
	Jugians	wainut	Ulmus americana	American eim
	Juglans cinera	butternut	Ulmus pumila	Siberian elm
	Juglans nigra	black walnut	Ulmus rubra	slippery elm
	Malus	apple spp.	Ulmus thomasii	rock elm
	Malua aaranaria	avve at anabanala	l halva avva	Unknown dead
	Malus coronaria	sweet crabappie	UNKNOWN	nardwood
Northorn Dine				
	Pinus banksiana	Jack pine	Pinus rigida	pitch pine
influence of SO4 on	Pinus nigra	Austrian pine	Pinus strobus	white pine
mortality	Pinus ponderosa	Ponderosa pine	Pinus sylvestris	Scotch pine
<ul> <li>negative influence of NO<sub>3</sub> on mortality</li> </ul>	Pinus resinosa	red pine		
	Asimina triloba	pawpaw	Morus	mulberry
	Carya alba	mockernut hickory	Nyssa sylvatica	blackgum
	Carya carolinae-	southern shagbark		oak spp
	septentrionalis	hickory	Quercus	Deciduous
Southern	Carya glabra	pignut hickory	Quercus buckleyi	Buckley oak
Midsuccessional	Carya illinoinensis	pecan	Quercus coccinia	scarlet oak
Hardwood	Carya laciniosa	shellbark hickory	Quercus falcata	southern red oak
<ul> <li>Large positive</li> </ul>	Carya myristiciformis	nutmeg hickory	Quercus imbricaria	shingle oak
influence of SO4	Carya pallida	sand hickory	Quercus incana	bluejack oak
deposition on	Carya texana	black hickory	Quercus laevis	turkey oak
mortality	Castanea mollissima	chinese chestnut	Quercus laurifolia	laurel oak
- negative milluence of NO₂ deposition	Castanea pumila	Chinkapin	Quercus margarettiae	runner oak
on mortality	Castanea pumila var			
,	ozarkensis	Ozark chinkapin	Quercus marilandica	blackjack oak
	Celtis	hackberry	Quercus michauxii	swamp chestnut oak
	Celtis laevigata	sugarberry	Quercus muehlenbergii	chinkapin oak
	Cercis canadensis	eastern redbud	Quercus nigra	water oak
	Cotinus obovatus	smoketree	Quercus oglethorpensis	Oglethorpe oak

Plant Functional Group	Genus Species	Common Name	Genus Species	Common Name
	Fraxinus	ash	Quercus pagoda	cherrybark oak
	Fraxinus caroliniana	Carolina ash	Quercus phellos	willow oak
	Fraxinus quadrangulata	blue ash	Quercus prinus	chestnut oak
	Liriodendron tulipifera	yellow-poplar	Quercus shumardii	Shumard's oak
	Magnolia	magnolia	Quercus similis	Delta post oak
	Magnolia acuminata	cucumbertree	Quercus sinuata var sinuata	Durand oak
	Magnolia fraseri	mountain magnolia	Quercus stellata	post oak
	Magnolia macrophylla	bigleaf magnolia	Triadica sebifera	Chinese tallowtree
	Magnolia tripetala	umbrella magnolia	Ulmus alata	winged elm
	Malus angustifolia	southern crabapple	Ulmus crassifolia	cedar elm
			Ulmus serotina	September elm
• ·· •	Pinus clausa	Sand pine		
Southern Pine	Pinus echinata	shortleaf pine		
- Large positive	Pinus elliottii	slash pine		
deposition on	Pinus glabra	spruce pine		
mortality	Pinus palustris	longleaf pine		
- negative influence	Pinus pungens	Table Mountain pine		
of NO <sub>3</sub> deposition	Pinus serotina	pond pine		
on mortality	Pinus taeda	loblolly pine		
	Pinus virginiana	Virginia pine		

1	
2	Attachment 2A
3	
4	Species-specific Sample Distribution across Ecoregions
5	for Species with Statistically Significant Associations of Growth with N/S
6	from Horn et al 2018 Supplemental Information Dataset
7	
8	Key:
9	NA_L2 = North American Ecoregion, code for level 2
10	NA_L3 = North American Ecoregion, code for level 3
11	US_L3NAME = Name of Ecoregion at level 3
12	See: <u>https://www.epa.gov/eco-research/ecoregions</u>
13	Median = Tree-specific median S and/or N deposition for the species samples
14	Assoc = U= unimodal, $\uparrow$ =positive, $\downarrow$ =negative
15	N/S = elation coefficient for N and S deposition values for the species samples
16	Count = number of species' tree samples assessed in all plots in that ecoregion
17	% = percent of species' tree samples in that ecoregion
18	
19	
16 17 18 19	% = percent of species' tree samples in that ecoregion

NA_ L2	NA_L3 CODE	US_L3NAME	boxe Media Asso N/S =	boxelder Median S=6 Assoc S-↓ N/S = 0.14		red maple Median N=9, S=7 Assoc N-↑, S-↓ N/S = 0.6		silver maple Median N=12,S=8 Assoc N-↑, S-↓ N/S = 0.27		yellow birch Median N=7 Assoc N-U N/S = 0.7		sweet birch Median S=12 Assoc S-↓ N/S = 0.58		<b>5irch</b> S=4 S-↓ 0.42
			count	%	count	%	count	%	count	%	count	%	count	%
	5.2.1	Northern Lakes and Forests	85	1.4%	23972	23.6%	324	7.1%	3282	23.9%			9247	50.1%
1	5.2.2	Northern Minnesota Wetlands	53	0.9%	93	0.1%			1	0.0%			547	3.0%
	5.3.1	Northeastern Highlands	17	0.3%	13245	13.1%	27	0.6%	6357	46.3%	1322	14.8%	4824	26.1%
1	5.3.3	North Central Appalachians			4883	4.8%			363	2.6%	1299	14.6%	93	0.5%
	6.2.3	Northern Rockies	1										129	0.7%
1	6.2.4	Canadian Rockies						_					6	0.0%
1	6.2.5	North Cascades											1	0.0%
1	6.2.7	Cascades											-	
1	6.2.8	Eastern Cascades Slopes and Foothills											-	
1	6.2.9	Blue Mountains											4	0.0%
1	6.2.10	Middle Rockies											16	0.1%
1	6.2.11	Klamath Mountains												
1	6.2.12	Sierra Nevada												
1	6.2.13	Wasatch and Uinta Mountains												
1	6.2.14	Southern Rockies												
1	6.2.15	Idaho Batholith											6	0.0%
	7.1.7	Puget Lowland											18	0.1%
1	7.1.8	Coast Range												
1	7.1.9	Willamette Valley											-	
	8.1.1	Eastern Great Lakes Lowlands	86	1.4%	1618	1.6%	293	6.4%	204	1.5%	49	0.6%	57	0.3%
ĺ	8.1.3	Northern Allegheny Plateau	8	0.1%	3565	3.5%	9	0.2%	394	2.9%	543	6.1%	82	0.4%
[	8.1.4	North Central Hardwood Forests	594	9.8%	4062	4.0%	448	9.8%	418	3.0%			978	5.3%

NA_ L2	NA_L3 CODE	US_L3NAME	$\begin{array}{c c} \textbf{boxelder} \\ \text{Median S=6} \\ \text{Assoc S-} \\ \text{N/S} = 0.14 \end{array} \begin{array}{c} \textbf{red maple} \\ \text{Median N=9,} \\ \text{Assoc N-} \\ \text{N/S} = 0.6 \end{array}$		red maple         silver maple         ye           ledian N=9, S=7         Median         Median         Median           Assoc N-↑, S-↓         N/S = 0.6         Assoc N-↑, S-↓         N/S = 0.27         N		yellow Media Assoc N/S	<b>birch</b> n N=7 c N-U = 0.7	sweet birch Median S=12 Assoc S-↓ N/S = 0.58		paper birch Median S=4 Assoc S-↓ N/S = 0.42			
			count	%	count	%	count	%	count	%	count	%	count	%
	8.1.5	Driftless Area	934	15.4%	631	0.6%	387	8.5%	31	0.2%			651	3.5%
1	8.1.6	Southern Michigan/Northern Indiana Drift Plains	106	1.7%	1463	1.4%	516	11.3%	36	0.3%			29	0.2%
1	8.1.7	Northeastern Coastal Zone	6	0.1%	4309	4.2%	18	0.4%	256	1.9%	975	10.9%	134	0.7%
1	8.1.8	Acadian Plains and Hills			5025	5.0%	9	0.2%	1236	9.0%			1437	7.8%
1	8.1.10	Erie Drift Plain	8	0.1%	1811	1.8%	83	1.8%	112	0.8%	6	0.1%		
	8.2.1	Southeastern Wisconsin Till Plains	338	5.6%	156	0.2%	124	2.7%	55	0.4%			55	0.3%
1	8.2.2	Huron/Erie Lake Plains	82	1.4%	1123	1.1%	195	4.3%	6	0.0%			123	0.7%
1	8.2.3	Central Corn Belt Plains	76	1.3%	13	0.0%	101	2.2%						
İ.	8.2.4	Eastern Corn Belt Plains	202	3.3%	333	0.3%	197	4.3%	1	0.0%				
	8.3.1	Northern Piedmont	71	1.2%	566	0.6%	38	0.8%	4	0.0%	109	1.2%		
1	8.3.2	Interior River Valleys and Hills	296	4.9%	423	0.4%	625	13.7%						
1	8.3.3	Interior Plateau	469	7.7%	1061	1.0%	82	1.8%						
1	8.3.4	Piedmont	135	2.2%	3119	3.1%					26	0.3%		
1	8.3.5	Southeastern Plains	154	2.5%	4363	4.3%	11	0.2%						
1	8.3.6	Mississippi Valley Loess Plains	192	3.2%	195	0.2%	35	0.8%						
1	8.3.7	South Central Plains	89	1.5%	742	0.7%	10	0.2%						
1	8.3.8	East Central Texas Plains	5	0.1%	4	0.0%		_						
	8.4.1	Ridge and Valley	100	1.6%	4942	4.9%	18	0.4%	166	1.2%	1866	21.0%	12	0.1%
1	8.4.2	Central Appalachians	17	0.3%	4912	4.8%	3	0.1%	495	3.6%	1170	13.1%		
1	8.4.3	Western Allegheny Plateau	193	3.2%	3926	3.9%	103	2.3%	17	0.1%	230	2.6%		
1	8.4.4	Blue Ridge	18	0.3%	3707	3.7%	3	0.1%	283	2.1%	1280	14.4%		
	8.4.5	Ozark Highlands	105	1.7%	185	0.2%	47	1.0%						

NA_ L2	NA_L3 CODE	US_L3NAME	boxe Media Asso N/S =	boxelder Median S=6 Assoc S-↓ N/S = 0.14		red maple Median N=9, S=7 Assoc N-↑, S-↓ N/S = 0.6		silver maple Median N=12,S=8 Assoc N-↑, S-↓ N/S = 0.27		yellow birch Median N=7 Assoc N-U N/S = 0.7		sweet birch Median S=12 Assoc S-↓ N/S = 0.58		<b>&gt;irch</b> S=4 S-↓ 0.42
			count	%	count	%	count	%	count	%	count	%	count	%
	8.4.6	Boston Mountains			174	0.2%								
1	8.4.7	Arkansas Valley	28	0.5%	56	0.1%	43	0.9%						
1	8.4.8	Ouachita Mountains	2	0.0%	156	0.2%	3	0.1%					-	
Î.	8.4.9	Southwestern Appalachians	25	0.4%	1401	1.4%			1	0.0%	24	0.3%		
	8.5.1	Middle Atlantic Coastal Plain	19	0.3%	2982	2.9%	15	0.3%						
	8.5.2	Mississippi Alluvial Plain	396	6.5%	471	0.5%	83	1.8%						
1	8.5.3	Southern Coastal Plain	4	0.1%	1425	1.4%								
i i	8.5.4	Atlantic Coastal Pine Barrens	1	0.0%	256	0.3%					6	0.1%		
	9.2.1	Northern Glaciated Plains	140	2.3%									4	0.0%
	9.2.2	Lake Agassiz Plain	200	3.3%	3	0.0%							6	0.0%
	9.2.3	Western Corn Belt Plains	555	9.1%	21	0.0%	420	9.2%	3	0.0%			6	0.0%
	9.2.4	Central Irregular Plains	157	2.6%	0	0.0%	273	6.0%						
	9.3.1	Northwestern Glaciated Plains	17	0.3%			2	0.0%						
İ.	9.3.3	Northwestern Great Plains	13	0.2%										1
1	9.3.4	Nebraska Sand Hills	1	0.0%										
	9.4.1	High Plains	3	0.0%										<u> </u>
Ì	9.4.2	Central Great Plains	48	0.8%			14	0.3%						
İ.	9.4.3	Southwestern Tablelands	4	0.1%										
İ.	9.4.4	Flint Hills	6	0.1%			1	0.0%						
1	9.4.5	Cross Timbers	3	0.0%			1	0.0%						
1	9.4.6	Edwards Plateau	1											
1	9.4.7	Texas Blackland Prairies												

NA_ L2	NA_L3 CODE	US_L3NAME	boxelder Median S=6 Assoc S-↓ N/S = 0.14		red maple Median N=9, S=7 Assoc N-↑, S-↓ N/S = 0.6		silver maple Median N=12,S=8 Assoc N-↑, S-↓ N/S = 0.27		yellow birch Median N=7 Assoc N-U N/S = 0.7		sweet birch Median S=12 Assoc S-↓ N/S = 0.58		paper birch Median S=4 Assoc S-↓ N/S = 0.42	
			count	%	count	%	count	%	count	%	count	%	count	%
	9.5.1	Western Gulf Coastal Plain			8	0.0%								
	9.6.1	Southern Texas Plains												
	10.1.2	Columbia Plateau												
1	10.1.3	Northern Basin and Range												
1	10.1.4	Wyoming Basin												
	10.1.5	Central Basin and Range												
1	10.1.6	Colorado Plateaus												
1	10.1.7	Arizona/New Mexico Plateau												
	10.1.8	Snake River Plain												
	10.2.1	Mojave Basin and Range												
	10.2.2	Sonoran Basin and Range												
	10.2.10	Chihuahuan Deserts												
	11.1.1	Southern and Central California Chaparral and Oak Woodlands												
1	11.1.2	Central California Valley	8	0.1%										
1	11.1.3	Southern California Mountains												
	12.1.1	Madrean Archipelago												
	13.1.1	Arizona/New Mexico Mountains	1	0.0%										
	15.4.1	Southern Florida Coastal Plain			34	0.0%								
		Total Tree Counts	6070		101434		4561		13721		8905		18465	

			Ame horn Media Asso N/S	erican Ibeam an S=7 oc S-↓ = 0.23	black h Median Assoc N/S =	<b>ickory</b> i N=10 ≳ N-↑ ÷ 0.17	hackl Mediar Assoc N/S =	<b>berry</b> n N=11 c N-U : 0.17	Amer bee Mec N=8, Assoc N N/S =	rican ech lian S=7 I-U,S-↓ ÷ 0.76	whit Media Asso N/S	<b>e ash</b> n N=10 bc N-↑ = 0.54	<b>green</b> Median N Assoc N- N/S =	<b>ash</b> =10, =6 ·U, S-↓ 0.45
L2	NA_L3 CODE	US L3NAME	count	%	count	%	count	%	count	%	count	%	count	%
	5.2.1	Northern Lakes and Forests	9	0.4%					1378	6.6%	1273	7.4%	1807	11.6%
	5.2.2	Northern Minnesota Wetlands					1	0.0%					200	1.3%
	5.3.1	Northeastern Highlands	9	0.4%					8502	40.7%	2438	14.1%	39	0.3%
İ	5.3.3	North Central Appalachians	45	2.1%					1520	7.3%	455	2.6%		
	6.2.3	Northern Rockies												
1	6.2.4	Canadian Rockies												
1	6.2.5	North Cascades												
İ	6.2.7	Cascades												
1	6.2.8	Eastern Cascades Slopes and Foothills												
Î.	6.2.9	Blue Mountains											_	
İ	6.2.10	Middle Rockies					1	0.0%					24	0.2%
1	6.2.11	Klamath Mountains											_	
1	6.2.12	Sierra Nevada												
	6.2.13	Wasatch and Uinta Mountains												
1	6.2.14	Southern Rockies												
İ	6.2.15	Idaho Batholith			<u> </u>								_	
	7.1.7	Puget Lowland											-	
İ	7.1.8	Coast Range												
1	7.1.9	Willamette Valley												
	8.1.1	Eastern Great Lakes Lowlands	7	0.3%			3	0.1%	282	1.3%	652	3.8%	479	3.1%
	8.1.3	Northern Allegheny Plateau	28	1.3%					1192	5.7%	1721	10.0%	76	0.5%

			Ame horn Media Asso N/S	erican Ibeam an S=7 bc S-↓ = 0.23	black h Median Assoc N/S =	i <b>ickory</b> า N=10 c N-↑ = 0.17	hackl Median Assoc N/S =	<b>berry</b> n N=11 c N-U = 0.17	Amei bee Mec N=8, Assoc N N/S =	' <b>ican</b> ⊭ <b>ch</b> Iian ,S=7 I-U,S-↓ = 0.76	white Mediai Asso N/S :	<b>e ash</b> n N=10 vc N-↑ = 0.54	<b>green</b> Median N: Assoc N- N/S =	<b>ash</b> =10, =6 U, S-↓ 0.45
INA_ L2	NA_L3	US L3NAME	count	%	count	%	count	%	count	%	count	%	count	%
	8.1.4	North Central Hardwood Forests	4	0.2%			46	0.9%	155	0.7%	595	3.4%	1429	9.2%
İ	8.1.5	Driftless Area	2	0.1%			235	4.8%	0	0.0%	354	2.1%	187	1.2%
	8.1.6	Southern Michigan/Northern Indiana Drift Plains	5	0.2%			35	0.7%	143	0.7%	265	1.5%	913	5.9%
	8.1.7	Northeastern Coastal Zone	2	0.1%			5	0.1%	327	1.6%	399	2.3%	58	0.4%
İ	8.1.8	Acadian Plains and Hills							1470	7.0%	758	4.4%	31	0.2%
	8.1.10	Erie Drift Plain	10	0.5%			3	0.1%	290	1.4%	465	2.7%	128	0.8%
	8.2.1	Southeastern Wisconsin Till Plains					23	0.5%	43	0.2%	177	1.0%	675	4.3%
	8.2.2	Huron/Erie Lake Plains	1	0.0%			13	0.3%	29	0.1%	84	0.5%	667	4.3%
	8.2.3	Central Corn Belt Plains	3	0.1%			75	1.5%			45	0.3%	102	0.7%
	8.2.4	Eastern Corn Belt Plains	9	0.4%			271	5.5%	110	0.5%	708	4.1%	286	1.8%
	8.3.1	Northern Piedmont	8	0.4%			29	0.6%	76	0.4%	256	1.5%	50	0.3%
	8.3.2	Interior River Valleys and Hills	13	0.6%	79	2.0%	591	12.1%	120	0.6%	477	2.8%	547	3.5%
	8.3.3	Interior Plateau	72	3.4%	24	0.6%	1031	21.0%	735	3.5%	1408	8.2%	714	4.6%
	8.3.4	Piedmont	252	11.8%			65	1.3%	521	2.5%	291	1.7%	481	3.1%
	8.3.5	Southeastern Plains	595	27.8%	7	0.2%	44	0.9%	609	2.9%	110	0.6%	1054	6.8%
Ì	8.3.6	Mississippi Valley Loess Plains	175	8.2%	26	0.7%	8	0.2%	102	0.5%	82	0.5%	254	1.6%
	8.3.7	South Central Plains	469	21.9%	190	4.8%	9	0.2%	152	0.7%	140	0.8%	561	3.6%
	8.3.8	East Central Texas Plains	2	0.1%	87	2.2%	2	0.0%			36	0.2%	168	1.1%
	8.4.1	Ridge and Valley	19	0.9%			138	2.8%	434	2.1%	909	5.3%	196	1.3%
Ì	8.4.2	Central Appalachians	36	1.7%			5	0.1%	1403	6.7%	408	2.4%	44	0.3%
	8.4.3	Western Allegheny Plateau	30	1.4%			70	1.4%	678	3.2%	1138	6.6%	106	0.7%
ľ	8.4.4	Blue Ridge	15	0.7%			5	0.1%	294	1.4%	317	1.8%	43	0.3%

			Ame horr Media Asso N/S	erican Ibeam an S=7 oc S-↓ = 0.23	black h Mediar Assoc N/S =	nickory n N=10 c N-↑ = 0.17	hackl Mediar Assoc N/S =	<b>berry</b> n N=11 c N-U = 0.17	Amer bee Mec N=8, Assoc N N/S =	<b>ican</b> <b>ch</b> ian S=7 I-U,S-↓ 0.76	whit Media Asso N/S ∹	<b>e ash</b> n N=10 c N-↑ = 0.54	<b>green</b> Median N Assoc N- N/S =	<b>ash</b> =10, =6 :U, S-↓ 0.45
L2	CODE	US L3NAME	count	%	count	%	count	%	count	%	count	%	count	%
	8.4.5	Ozark Highlands	5	0.2%	1863	46.6%	262	5.3%			558	3.2%	155	1.0%
1	8.4.6	Boston Mountains	4	0.2%	681	17.0%	20	0.4%	55	0.3%	79	0.5%	18	0.1%
1	8.4.7	Arkansas Valley	10	0.5%	576	14.4%	32	0.7%			65	0.4%	137	0.9%
1	8.4.8	Ouachita Mountains	24	1.1%	385	9.6%	2	0.0%	8	0.0%	23	0.1%	80	0.5%
1	8.4.9	Southwestern Appalachians	27	1.3%	1	0.0%	19	0.4%	152	0.7%	214	1.2%	119	0.8%
	8.5.1	Middle Atlantic Coastal Plain	101	4.7%			21	0.4%	89	0.4%	34	0.2%	369	2.4%
1	8.5.2	Mississippi Alluvial Plain	28	1.3%	21	0.5%	55	1.1%	9	0.0%	9	0.1%	717	4.6%
1	8.5.3	Southern Coastal Plain	108	5.1%			9	0.2%	2	0.0%	6	0.0%	440	2.8%
1	8.5.4	Atlantic Coastal Pine Barrens	2	0.1%			0	0.0%	14	0.1%	7	0.0%		
	9.2.1	Northern Glaciated Plains					14	0.3%					337	2.2%
1	9.2.2	Lake Agassiz Plain											254	1.6%
1	9.2.3	Western Corn Belt Plains					571	11.6%			82	0.5%	416	2.7%
1	9.2.4	Central Irregular Plains			45	1.1%	779	15.9%			216	1.3%	354	2.3%
	9.3.1	Northwestern Glaciated Plains					14	0.3%					81	0.5%
1	9.3.3	Northwestern Great Plains					3	0.1%					360	2.3%
1	9.3.4	Nebraska Sand Hills					6	0.1%					25	0.2%
	9.4.1	High Plains					2	0.0%					18	0.1%
1	9.4.2	Central Great Plains					235	4.8%			1	0.0%	268	1.7%
1	9.4.3	Southwestern Tablelands					10	0.2%					9	0.1%
1	9.4.4	Flint Hills					131	2.7%			1	0.0%	42	0.3%
ĺ	9.4.5	Cross Timbers			12	0.3%	9	0.2%			7	0.0%	24	0.2%
[	9.4.6	Edwards Plateau												

			Ame horn Media Asso N/S	erican beam an S=7 oc S-↓ = 0.23	black h Median Assoc N/S =	<b>iickory</b> n N=10 c N-↑ : 0.17	hackl Median Assoc N/S =	<b>berry</b> n N=11 c N-U = 0.17	Amer bee Med N=8, Assoc N N/S =	<b>ican</b> <b>ch</b> ian S=7 I-U,S-↓ 0.76	<b>whit</b> e Mediai Asso N/S =	<b>e ash</b> n N=10 c N-↑ = 0.54	<b>green</b> Median N: Assoc N- N/S =	<b>ash</b> =10, =6 U, S-↓ 0.45
L2	CODE	US L3NAME	count	%	count	%	count	%	count	%	count	%	count	%
	9.4.7	Texas Blackland Prairies									3	0.0%		
	9.5.1	Western Gulf Coastal Plain	8	0.4%									28	0.2%
	9.6.1	Southern Texas Plains												
	10.1.2	Columbia Plateau												
Ì	10.1.3	Northern Basin and Range												
1	10.1.4	Wyoming Basin												
1	10.1.5	Central Basin and Range												
İ	10.1.6	Colorado Plateaus												
1	10.1.7	Arizona/New Mexico Plateau												
1	10.1.8	Snake River Plain												
	10.2.1	Mojave Basin and Range												
	10.2.2	Sonoran Basin and Range												
	10.2.10	Chihuahuan Deserts												
	11.1.1	Southern and Central California Chaparral and Oak Woodlands												
1	11.1.2	Central California Valley												
1	11.1.3	Southern California Mountains												
	12.1.1	Madrean Archipelago												
	13.1.1	Arizona/New Mexico Mountains												
	15.4.1	Southern Florida Coastal Plain											3	0.0%
		Total Tree Counts	2137		3997		4902		20894		17266		15573	

NA	NA 12		honey Media Asso N/S	<b>/locust</b> an S=6 ic S-↓ = 0.27	black v Med N=12 Assoc N N/S =	<b>walnut</b> dian 2,S=9 √-↑, S-↓ = 0.08	Utah ji Median N Assoc N N/S =	<b>uniper</b> √=3, S=1 I-↑, S-↓ = 0.71	east redc Media Assoc N/S	tern edar n S=7 c S-↓ = 0.3	swee Median Assoc I N/S	e <b>tgum</b> N=9, S=7 N-↑, S-↓ = 0.37	<b>yellow-p</b> Median Assoc N/S =	<b>xoplar</b> N=10 N-↑ 0.41
L2	CODE	US L3NAME	count	%	count	%	count	%	count	%	count	%	count	%
	5.2.1	Northern Lakes and Forests				1			2	0.0%				
1	5.2.2	Northern Minnesota Wetlands									-			
	5.3.1	Northeastern Highlands			6	0.1%			16	0.1%			91	0.4%
	5.3.3	North Central Appalachians							4	0.0%			47	0.2%
	6.2.3	Northern Rockies												+
	6.2.4	Canadian Rockies			-									
1	6.2.5	North Cascades												
	6.2.7	Cascades												
1	6.2.8	Eastern Cascades Slopes and Foothills												+
	6.2.9	Blue Mountains												+
	6.2.10	Middle Rockies					33	0.3%						+
1	6.2.11	Klamath Mountains												
1	6.2.12	Sierra Nevada												
1	6.2.13	Wasatch and Uinta Mountains					698	6.3%						
1	6.2.14	Southern Rockies					110	1.0%						
	6.2.15	Idaho Batholith												
	7.1.7	Puget Lowland												
	7.1.8	Coast Range												+
1	7.1.9	Willamette Valley												
	8.1.1	Eastern Great Lakes Lowlands			30	0.5%			29	0.2%				1
1	8.1.3	Northern Allegheny Plateau			34	0.6%			11	0.1%				1

			honey Media Asso N/S	<b>/locust</b> an S=6 bc S-↓ = 0.27	black w Med N=12 Assoc N N/S =	<b>valnut</b> lian ∖,S=9 I-↑, S-↓ = 0.08	Utah ju Median N Assoc N N/S =	<b>ıniper</b> I=3, S=1 I-↑, S-↓ ∺ 0.71	east redc Media Asso N/S	t <b>ern edar</b> In S=7 c S-↓ = 0.3	swee Median I Assoc I N/S	<b>؛tgum</b> N=9, S=7 N-↑, S-↓ = 0.37	<b>yellow-p</b> Median Assoc N/S =	r <b>oplar</b> N=10 N-↑ 0.41
NA_ L2	CODE	US L3NAME	count	%	count	%	count	%	count	%	count	%	count	%
	8.1.4	North Central Hardwood Forests			13	0.2%			109	0.8%		1		
	8.1.5	Driftless Area	6	0.3%	404	7.1%			276	1.9%				
	8.1.6	Southern Michigan/Northern Indiana Drift Plains	2	0.1%	119	2.1%			24	0.2%			58	0.2%
	8.1.7	Northeastern Coastal Zone			14	0.2%			84	0.6%	2	0.0%	46	0.2%
	8.1.8	Acadian Plains and Hills			1					1 1				1
	8.1.10	Erie Drift Plain	1	0.0%	50	0.9%				1 1	1	0.0%	160	0.7%
	8.2.1	Southeastern Wisconsin Till Plains	1	0.0%	80	1.4%			88	0.6%		+		
	8.2.2	Huron/Erie Lake Plains	7	0.3%	35	0.6%			1	0.0%		+	4	0.0%
	8.2.3	Central Corn Belt Plains	72	3.6%	130	2.3%			8	0.1%		+	3	0.0%
	8.2.4	Eastern Corn Belt Plains	130	6.5%	417	7.4%			142	1.0%	62	0.2%	183	0.8%
	8.3.1	Northern Piedmont	1	0.0%	142	2.5%			221	1.5%	37	0.1%	659	2.7%
	8.3.2	Interior River Valleys and Hills	203	10.1%	457	8.1%			622	4.3%	444	1.5%	377	1.6%
	8.3.3	Interior Plateau	166	8.3%	796	14.0%			3325	23.1%	791	2.7%	2259	9.3%
	8.3.4	Piedmont	13	0.6%	153	2.7%			1031	7.2%	5544	19.0%	5178	21.4%
	8.3.5	Southeastern Plains	11	0.5%	54	1.0%			645	4.5%	9331	32.0%	3421	14.2%
	8.3.6	Mississippi Valley Loess Plains	32	1.6%	24	0.4%			163	1.1%	1538	5.3%	272	1.1%
	8.3.7	South Central Plains	58	2.9%	14	0.2%			167	1.2%	4762	16.3%	13	0.1%
	8.3.8	East Central Texas Plains	17	0.8%	4	0.1%			122	0.8%	209	0.7%		
	8.4.1	Ridge and Valley	10	0.5%	353	6.2%			722	5.0%	546	1.9%	1657	6.9%
	8.4.2	Central Appalachians	4	0.2%	50	0.9%			41	0.3%	94	0.3%	2997	12.4%
	8.4.3	Western Allegheny Plateau	15	0.7%	386	6.8%			44	0.3%	14	0.0%	2390	9.9%
	8.4.4	Blue Ridge	6	0.3%	65	1.1%			32	0.2%	65	0.2%	2779	11.5%

			honey Media Asso N/S	<b>/locust</b> an S=6 oc S-↓ = 0.27	black v Mec N=12 Assoc N N/S =	<b>walnut</b> dian 2,S=9 I-↑, S-↓ = 0.08	Utah ju Median N Assoc N N/S =	<b>uniper</b> I=3, S=1 I-↑, S-↓ © 0.71	east redc Media Asso N/S	tern edar n S=7 c S-↓ = 0.3	swee Median Assoc I N/S	etgum N=9, S=7 N-↑, S-↓ = 0.37	<b>yellow-</b> Median Assoc N/S =	ooplar N=10 N-↑ 0.41
L2	CODE	US L3NAME	count	%	count	%	count	%	count	%	count	%	count	%
	8.4.5	Ozark Highlands	180	9.0%	710	12.5%			3519	24.5%	146	0.5%	4	0.0%
	8.4.6	Boston Mountains	6	0.3%	28	0.5%			285	2.0%	172	0.6%		
	8.4.7	Arkansas Valley	15	0.7%	9	0.2%			606	4.2%	226	0.8%		
	8.4.8	Ouachita Mountains	17	0.8%	4	0.1%			210	1.5%	311	1.1%		
	8.4.9	Southwestern Appalachians	4	0.2%	37	0.7%			370	2.6%	620	2.1%	1035	4.3%
	8.5.1	Middle Atlantic Coastal Plain	1	0.0%	12	0.2%			13	0.1%	2428	8.3%	467	1.9%
1	8.5.2	Mississippi Alluvial Plain	100	5.0%	7	0.1%			19	0.1%	574	2.0%	10	0.0%
1	8.5.3	Southern Coastal Plain	2	0.1%					26	0.2%	1111	3.8%	43	0.2%
	8.5.4	Atlantic Coastal Pine Barrens			2	0.0%			10	0.1%	60	0.2%	16	0.1%
	9.2.1	Northern Glaciated Plains	9	0.4%										
	9.2.2	Lake Agassiz Plain												
i i	9.2.3	Western Corn Belt Plains	345	17.2%	325	5.7%			318	2.2%				
İ	9.2.4	Central Irregular Plains	496	24.7%	617	10.9%			381	2.6%				
	9.3.1	Northwestern Glaciated Plains							118	0.8%				
1	9.3.3	Northwestern Great Plains			1	0.0%			82	0.6%				
1	9.3.4	Nebraska Sand Hills							92	0.6%				
	9.4.1	High Plains			1	0.0%			23	0.2%				
	9.4.2	Central Great Plains	46	2.3%	22	0.4%			275	1.9%				
1	9.4.3	Southwestern Tablelands			6	0.1%			8	0.1%				
	9.4.4	Flint Hills	30	1.5%	45	0.8%			55	0.4%				
1	9.4.5	Cross Timbers	3	0.1%	10	0.2%			17	0.1%				
[	9.4.6	Edwards Plateau		<u> </u>										

NA			hone Medi Asso N/S	<b>ylocust</b> an S=6 oc S-↓ = 0.27	black w Med N=12 Assoc N N/S =	<b>valnut</b> lian ,S=9 l-↑, S-↓ : 0.08	Utah ju Median N Assoc N N/S =	u <b>niper</b> √=3, S=1 I-↑, S-↓ ፥ 0.71	east redco Mediar Assoc N/S =	ern edar n S=7 c S-↓ = 0.3	swee Median Assoc I N/S	e <b>tgum</b> N=9, S=7 N-↑, S-↓ = 0.37	yellow-p Median I Assoc I N/S = (	o <b>plar</b> N=10 N-↑ 0.41
L2	CODE	US_L3NAME	count	%	count	%	count	%	count	%	count	%	count	%
	9.4.7	Texas Blackland Prairies							12	0.1%				
	9.5.1	Western Gulf Coastal Plain							11	0.1%	92	0.3%		
	9.6.1	Southern Texas Plains												
	10.1.2	Columbia Plateau												
	10.1.3	Northern Basin and Range					405	3.7%						
	10.1.4	Wyoming Basin					66	0.6%					-	
	10.1.5	Central Basin and Range					3112	28.1%						
	10.1.6	Colorado Plateaus					3935	35.5%					-	
	10.1.7	Arizona/New Mexico Plateau					1601	14.4%						
	10.1.8	Snake River Plain												
	10.2.1	Mojave Basin and Range					115	1.0%						
	10.2.2	Sonoran Basin and Range											-	
	10.2.10	Chihuahuan Deserts												
	11.1.1	Southern and Central California Chaparral and Oak Woodlands												
	11.1.2	Central California Valley												
	11.1.3	Southern California Mountains												
	12.1.1	Madrean Archipelago					1	0.0%						
	13.1.1	Arizona/New Mexico Mountains					1008	9.1%						
	15.4.1	Southern Florida Coastal Plain												
		Total Tree Count	2009		5666		11084		14379		29180		24169	

			tar Media Asso N/S	<b>noak</b> an N=4 bc N-U = 0.57	Osage- Mediai Assoc N/S =	orange n S=5 c S-↓ : 0.36	swee Mediai Assoc N/S =	<b>tbay</b> n N=7 : N-U : 0.34	water t Mediar Assoc N/S =	<b>upelo</b> n S=8 : S-↓ = 0.5	swamp Media Asso N/S	o <b>tupelo</b> an N=7 c N-U = 0.47	white s Mediar Assoc N/S	<b>pruce</b> ı S=4 : S-↓ =
NA_ L2	NA_L3 CODE	US L3NAME	count	%	count	%	count	%	count	%	count	%	count	%
	5.2.1	Northern Lakes and Forests											3739	63.0%
	5.2.2	Northern Minnesota Wetlands											245	4.1%
	5.3.1	Northeastern Highlands											716	12.1%
	5.3.3	North Central Appalachians											28	0.5%
	6.2.3	Northern Rockies												+
	6.2.4	Canadian Rockies												1
	6.2.5	North Cascades												-
	6.2.7	Cascades	1	0.0%										-
	6.2.8	Eastern Cascades Slopes and Foothills												-
	6.2.9	Blue Mountains												-
	6.2.10	Middle Rockies											194	3.3%
	6.2.11	Klamath Mountains	1561	51.9%										-
	6.2.12	Sierra Nevada	116	3.9%										1
	6.2.13	Wasatch and Uinta Mountains												
	6.2.14	Southern Rockies												
	6.2.15	Idaho Batholith												1
	7.1.7	Puget Lowland												
	7.1.8	Coast Range	1276	42.4%										
	7.1.9	Willamette Valley												+
	8.1.1	Eastern Great Lakes Lowlands											2	0.0%
	8.1.3	Northern Allegheny Plateau											7	0.1%

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			tan Media Asso N/S	<b>ioak</b> an N=4 bc N-U = 0.57	Osage- Medial Assoc N/S =	<b>orange</b> n S=5 c S-↓ : 0.36	swee Media Assoc N/S =	e <b>tbay</b> n N=7 c N-U = 0.34	water Media Asso N/S	<b>tupelo</b> n S=8 c S-↓ = 0.5	swamp Media Asso N/S	<b>tupelo</b> In N=7 c N-U = 0.47	white s Median Assoc N/S =	oruce S=4 S-↓
NA_ L2	NA_L3 CODE	US L3NAME	count	%	count	%	count	%	count	%	count	%	count	%
	8.1.4	North Central Hardwood Forests											127	2.1%
1	8.1.5	Driftless Area											43	0.7%
1	8.1.6	Southern Michigan/Northern Indiana Drift Plains			3	0.1%							30	0.5%
1	8.1.7	Northeastern Coastal Zone											2	0.0%
1	8.1.8	Acadian Plains and Hills											754	12.7%
1	8.1.10	Erie Drift Plain			17	0.7%								
	8.2.1	Southeastern Wisconsin Till Plains											25	0.4%
1	8.2.2	Huron/Erie Lake Plains											2	0.0%
	8.2.3	Central Corn Belt Plains			73	3.1%								
	8.2.4	Eastern Corn Belt Plains			139	5.8%								
	8.3.1	Northern Piedmont			8	0.3%							1	0.0%
ľ	8.3.2	Interior River Valleys and Hills			231	9.7%			33	1.3%				
1	8.3.3	Interior Plateau			281	11.8%			2	0.1%	1	0.0%		
1	8.3.4	Piedmont			2	0.1%	34	1.0%			56	0.7%		
1	8.3.5	Southeastern Plains			72	3.0%	1848	56.6%	686	26.3%	3615	45.6%		
1	8.3.6	Mississippi Valley Loess Plains			5	0.2%	11	0.3%	59	2.3%	5	0.1%		
1	8.3.7	South Central Plains			81	3.4%	188	5.8%	147	5.6%	43	0.5%		
İ.	8.3.8	East Central Texas Plains			47	2.0%								
	8.4.1	Ridge and Valley			49	2.1%	2	0.1%			1	0.0%		
ĺ	8.4.2	Central Appalachians												
	8.4.3	Western Allegheny Plateau			57	2.4%							4	0.1%
	8.4.4	Blue Ridge												1

			tan Media Asso N/S	o <b>ak</b> an N=4 c N-U = 0.57	Osage- Media Assoc N/S =	<b>orange</b> n S=5 c S-↓ = 0.36	swee Media Assoc N/S =	e <b>tbay</b> n N=7 c N-U = 0.34	water Media Asso N/S	<b>tupelo</b> n S=8 c S-↓ = 0.5	<b>swamı</b> Media Asso N/S	<b>o tupelo</b> an N=7 oc N-U = 0.47	white s Median Assoc N/S =	oruce S=4 S-↓
NA_ L2	NA_L3 CODE	US L3NAME	count	%	count	%	count	%	count	%	count	%	count	%
	8.4.5	Ozark Highlands			105	4.4%								
1	8.4.6	Boston Mountains			1	0.0%								
1	8.4.7	Arkansas Valley			16	0.7%					2	0.0%		
1	8.4.8	Ouachita Mountains			24	1.0%								
1	8.4.9	Southwestern Appalachians							2	0.1%				
	8.5.1	Middle Atlantic Coastal Plain					170	5.2%	540	20.7%	1491	18.8%		
	8.5.2	Mississippi Alluvial Plain							682	26.2%	24	0.3%		
İ.	8.5.3	Southern Coastal Plain			·		993	30.4%	391	15.0%	2697	34.0%		
	8.5.4	Atlantic Coastal Pine Barrens					10	0.3%						
	9.2.1	Northern Glaciated Plains												
1	9.2.2	Lake Agassiz Plain											4	0.1%
1	9.2.3	Western Corn Belt Plains			141	5.9%							12	0.2%
1	9.2.4	Central Irregular Plains			758	31.8%								
	9.3.1	Northwestern Glaciated Plains												
	9.3.3	Northwestern Great Plains												
1	9.3.4	Nebraska Sand Hills												
	9.4.1	High Plains												
1	9.4.2	Central Great Plains			88	3.7%								
	9.4.3	Southwestern Tablelands			7	0.3%								
1	9.4.4	Flint Hills			154	6.5%								
1	9.4.5	Cross Timbers			22	0.9%								
[	9.4.6	Edwards Plateau				1								

			tar Media Asso N/S	noak an N=4 bc N-U = 0.57	Osage- Median Assoc N/S =	orange n S=5 c S-↓ : 0.36	swee Media Assoc N/S =	<b>tbay</b> n N=7 : N-U : 0.34	water f Mediar Assoc N/S =	t <b>upelo</b> n S=8 c S-↓ = 0.5	swamp Media Asso N/S :	<b>tupelo</b> in N=7 c N-U = 0.47	white sp Median Assoc N/S =	S=4 S-↓
NA_ L2	NA_L3	US_L3NAME	count	%	count	%	count	%	count	%	count	%	count	%
	9.4.7	Texas Blackland Prairies			1	0.0%								
	9.5.1	Western Gulf Coastal Plain			2	0.1%	1	0.0%	65	2.5%				
	9.6.1	Southern Texas Plains												
	10.1.2	Columbia Plateau												
	10.1.3	Northern Basin and Range												
	10.1.4	Wyoming Basin												
	10.1.5	Central Basin and Range												
	10.1.6	Colorado Plateaus												
	10.1.7	Arizona/New Mexico Plateau					-							
	10.1.8	Snake River Plain												
	10.2.1	Mojave Basin and Range												
	10.2.2	Sonoran Basin and Range												
	10.2.10	Chihuahuan Deserts					-							
	11.1.1	Southern and Central California Chaparral and Oak Woodlands	55	1.8%										
	11.1.2	Central California Valley												
	11.1.3	Southern California Mountains												
	12.1.1	Madrean Archipelago												
	13.1.1	Arizona/New Mexico Mountains												
	15.4.1	Southern Florida Coastal Plain					6	0.2%			1	0.0%		
		Total Tree Count	3009		2384		3263		2607		7936		5935	

	NA_ NA_L3 L2 CODE		shortle Media Asso N/S	eafpine an S=6 ic S-↓ = 0.16	slash Media Assoc N/S =	<b>pine</b> n S=5 c S-↓ : 0.46	single piny Mediar Assoc N/S =	<b>eleaf</b> /on n N=3 c N-↑ : 0.58	longlea Mediar Assoc N/S =	<b>If pine</b> n N=8 : N-U 0.45	red Median Assoc I N/S	<b>pine</b> N=8, S=5 N-↑, S-↓ = 0.53	pitch   Median Assoc N/S =	p <b>ine</b> N=10 N-U 0.66
L2	CODE	US L3NAME	count	%	count	%	count	%	count	%	count	%	count	%
	5.2.1	Northern Lakes and Forests									5823	65.3%		
	5.2.2	Northern Minnesota Wetlands									192	2.2%		
	5.3.1	Northeastern Highlands									151	1.7%	81	3.1%
	5.3.3	North Central Appalachians									35	0.4%	37	1.4%
	6.2.3	Northern Rockies											1	
	6.2.4	Canadian Rockies												1
	6.2.5	North Cascades												-
	6.2.7	Cascades												-
	6.2.8	Eastern Cascades Slopes and Foothills												
	6.2.9	Blue Mountains												
	6.2.10	Middle Rockies												
	6.2.11	Klamath Mountains												
	6.2.12	Sierra Nevada					109	3.0%						
	6.2.13	Wasatch and Uinta Mountains					11	0.3%						
	6.2.14	Southern Rockies												
1	6.2.15	Idaho Batholith												
	7.1.7	Puget Lowland												
	7.1.8	Coast Range												-
	7.1.9	Willamette Valley												
	8.1.1	Eastern Great Lakes Lowlands									66	0.7%	6	0.2%
	8.1.3	Northern Allegheny Plateau									120	1.3%	2	0.1%

			shortle Media Asso N/S	<b>eaf pine</b> an S=6 oc S-↓ = 0.16	slash Media Assoc N/S =	<b>pine</b> n S=5 c S-↓ ≎ 0.46	single piny Mediar Assoc N/S =	eleaf /on n N=3 c N-↑ 0.58	longle: Media Assoc N/S =	<b>af pine</b> n N=8 c N-U = 0.45	red Median I Assoc I N/S	<b>pine</b> N=8, S=5 N-↑, S-↓ = 0.53	<b>pitch p</b> Median Assoc N/S =	<b>&gt;ine</b> N=10 N-U 0.66
L2	CODE	US L3NAME	count	%	count	%	count	%	count	%	count	%	count	%
	8.1.4	North Central Hardwood Forests									1456	16.3%		
1	8.1.5	Driftless Area									327	3.7%		
1	8.1.6	Southern Michigan/Northern Indiana Drift Plains									367	4.1%	1	0.0%
1	8.1.7	Northeastern Coastal Zone									7	0.1%	88	3.4%
1	8.1.8	Acadian Plains and Hills									159	1.8%	3	0.1%
İ.	8.1.10	Erie Drift Plain									12	0.1%		
	8.2.1	Southeastern Wisconsin Till Plains									80	0.9%		<u> </u>
i i	8.2.2	Huron/Erie Lake Plains									51	0.6%		
1	8.2.3	Central Corn Belt Plains									5	0.1%		
1	8.2.4	Eastern Corn Belt Plains									6	0.1%		
	8.3.1	Northern Piedmont	7	0.1%										<u> </u>
İ.	8.3.2	Interior River Valleys and Hills	19	0.1%							21	0.2%	2	0.1%
İ.	8.3.3	Interior Plateau	191	1.4%							2	0.0%	5	0.2%
1	8.3.4	Piedmont	2037	15.3%	16	0.2%			203	4.4%			17	0.7%
1	8.3.5	Southeastern Plains	1451	10.9%	3418	34.4%			2729	58.9%			1	0.0%
1	8.3.6	Mississippi Valley Loess Plains	94	0.7%	2	0.0%			1	0.0%				
1	8.3.7	South Central Plains	1336	10.1%	190	1.9%			244	5.3%				
ľ	8.3.8	East Central Texas Plains	23	0.2%	6	0.1%								
	8.4.1	Ridge and Valley	291	2.2%					87	1.9%	6	0.1%	363	14.1%
1	8.4.2	Central Appalachians	23	0.2%							11	0.1%	50	1.9%
1	8.4.3	Western Allegheny Plateau	39	0.3%							18	0.2%	81	3.1%
	8.4.4	Blue Ridge	275	2.1%									210	8.1%

NA	NA 12		shortle Media Asso N/S	eafpine an S=6 bc S-↓ = 0.16	slash Media Asso N/S =	n <b>pine</b> n S=5 c S-↓ = 0.46	single piny Mediar Assoc N/S =	eleaf /on n N=3 c N-↑ 0.58	Iongle Media Asso N/S :	<b>af pine</b> in N=8 c N-U = 0.45	red Median I Assoc I N/S	<b>pine</b> N=8, S=5 N-↑, S-↓ = 0.53	pitch Median Assoc N/S =	<b>pine</b> N=10 N-U 0.66
L2	CODE	US L3NAME	count	%	count	%	count	%	count	%	count	%	count	%
	8.4.5	Ozark Highlands	2400	18.1%										
1	8.4.6	Boston Mountains	567	4.3%										
1	8.4.7	Arkansas Valley	1059	8.0%										
	8.4.8	Ouachita Mountains	3137	23.6%										
1	8.4.9	Southwestern Appalachians	251	1.9%					9	0.2%			9	0.3%
	8.5.1	Middle Atlantic Coastal Plain	25	0.2%	105	1.1%			301	6.5%			33	1.3%
	8.5.2	Mississippi Alluvial Plain	11	0.1%										
1	8.5.3	Southern Coastal Plain	2	0.0%	6030	60.6%			1054	22.7%				
	8.5.4	Atlantic Coastal Pine Barrens	38	0.3%									1589	61.6%
	9.2.1	Northern Glaciated Plains												
1	9.2.2	Lake Agassiz Plain												
1	9.2.3	Western Corn Belt Plains												
1	9.2.4	Central Irregular Plains									2	0.0%		
	9.3.1	Northwestern Glaciated Plains												
1	9.3.3	Northwestern Great Plains												
1	9.3.4	Nebraska Sand Hills												
	9.4.1	High Plains												
	9.4.2	Central Great Plains												
1	9.4.3	Southwestern Tablelands												
1	9.4.4	Flint Hills												1
1	9.4.5	Cross Timbers	2	0.0%										
[	9.4.6	Edwards Plateau												

	NA 12		shortle Media Asso N/S	eafpine an S=6 bc S-↓ = 0.16	slash Mediar Assoc N/S =	<b>pine</b> n S=5 c S-↓ ≎ 0.46	singl ping Media Asso N/S =	<b>eleaf</b> yon n N=3 c N-↑ = 0.58	longlea Mediar Assoc N/S =	<b>af pine</b> n N=8 c N-U : 0.45	red Median I Assoc I N/S	<b>pine</b> N=8, S=5 N-↑, S-↓ = 0.53	<b>pitch p</b> Median I Assoc I N/S = (	v <b>ine</b> N=10 N-U 0.66
L2	CODE	US_L3NAME	count	%	count	%	count	%	count	%	count	%	count	%
	9.4.7	Texas Blackland Prairies												
	9.5.1	Western Gulf Coastal Plain							7	0.2%				
	9.6.1	Southern Texas Plains												
	10.1.2	Columbia Plateau												
	10.1.3	Northern Basin and Range					42	1.2%						
1	10.1.4	Wyoming Basin												
	10.1.5	Central Basin and Range					2988	83.5%						
1	10.1.6	Colorado Plateaus					46	1.3%					-	
	10.1.7	Arizona/New Mexico Plateau					86	2.4%						
1	10.1.8	Snake River Plain												
	10.2.1	Mojave Basin and Range					153	4.3%						
	10.2.2	Sonoran Basin and Range												
	10.2.10	Chihuahuan Deserts												
	11.1.1	Southern and Central California Chaparral and Oak Woodlands												
	11.1.2	Central California Valley												
	11.1.3	Southern California Mountains					127	3.5%						
	12.1.1	Madrean Archipelago					3	0.1%						
	13.1.1	Arizona/New Mexico Mountains					14	0.4%						
	15.4.1	Southern Florida Coastal Plain			178	1.8%								
		Total Tree Coun			9945		3579		4635		8917		2578	

			easter p Median Assoc N/S	<b>'n white</b> ine N=8, S=6 N-↑, S-↓ = 0.59	Iobloll Media Asso N/S	<b>y pine</b> n S=7 c S-↓ = 0.32	bigtooth Media Asso N/S =	n <b>aspen</b> n S=6 c S-↓ = 0.57	quak asp Media Assor N/S	<b>(ing</b> <b>)en</b> n N=7 c N-U = 0.6	black Media Assc N/S	<b>cherry</b> n N=11 c N-U = 0.33	Dougla Median N Assoc N- N/S =	<b>₄s-fir</b> =3, S=1 -↑, S-↓ 0.65
L2	CODE	US L3NAME	count	%	count	%	count	%	count	%	count	%	count	%
	5.2.1	Northern Lakes and Forests	3921	19.2%			6123	61.0%	23006	55.1%	1726	8.4%		1
	5.2.2	Northern Minnesota Wetlands	34	0.2%			13	0.1%	2488	6.0%			-	1
	5.3.1	Northeastern Highlands	3744	18.3%		1	397	4.0%	961	2.3%	1284	6.3%	-	1
	5.3.3	North Central Appalachians	525	2.6%			89	0.9%	136	0.3%	1260	6.2%	-	1
	6.2.3	Northern Rockies		1			1	1	44	0.1%			4096	10.4%
	6.2.4	Canadian Rockies							74	0.2%		1	627	1.6%
	6.2.5	North Cascades								1		1	2101	5.3%
	6.2.7	Cascades				1		1				1	8882	22.6%
	6.2.8	Eastern Cascades Slopes and Foothills							20	0.0%		1	1394	3.5%
	6.2.9	Blue Mountains							6	0.0%		1	2946	7.5%
	6.2.10	Middle Rockies							264	0.6%			3404	8.6%
	6.2.11	Klamath Mountains							3	0.0%		1	5771	14.7%
	6.2.12	Sierra Nevada							21	0.1%			880	2.2%
	6.2.13	Wasatch and Uinta Mountains							2195	5.3%			653	1.7%
	6.2.14	Southern Rockies							3606	8.6%		1	1841	4.7%
	6.2.15	Idaho Batholith							3	0.0%			1294	3.3%
	7.1.7	Puget Lowland								++		1	335	0.9%
	7.1.8	Coast Range				1		1				1	3526	9.0%
	7.1.9	Willamette Valley								1		1	155	0.4%
	8.1.1	Eastern Great Lakes Lowlands	515	2.5%			71	0.7%	266	0.6%	344	1.7%		1
	8.1.3	Northern Allegheny Plateau	769	3.8%			138	1.4%	340	0.8%	697	3.4%	8	0.0%

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	NA 12		easter p Median Assoc N/S	r <b>n white</b> ine N=8, S=6 N-↑, S-↓ = 0.59	<b>lobioli</b> Media Assoc N/S =	<b>y pine</b> n S=7 c S-↓ = 0.32	bigtooth Median Assoc N/S =	n <b>aspen</b> n S=6 c S-↓ : 0.57	qual asp Mediar Assoc N/S :	<b>king</b> n N=7 n N-U = 0.6	black Media Asso N/S	<b>cherry</b> n N=11 ic N-U = 0.33	Dougla Median N Assoc N- N/S =	<b>ìs-fir</b> =3, S=1 ·↑, S-↓ 0.65
L2	CODE	US_L3NAME	count	%	count	%	count	%	count	%	count	%	count	%
	8.1.4	North Central Hardwood Forests	1587	7.8%			935	9.3%	3258	7.8%	608	3.0%		
	8.1.5	Driftless Area	379	1.9%			425	4.2%	383	0.9%	774	3.8%		
	8.1.6	Southern Michigan/Northern Indiana Drift Plains	320	1.6%	4	0.0%	308	3.1%	250	0.6%	1233	6.0%	1	0.0%
	8.1.7	Northeastern Coastal Zone	2299	11.2%			124	1.2%	152	0.4%	244	1.2%		1
1	8.1.8	Acadian Plains and Hills	2113	10.3%			513	5.1%	970	2.3%	136	0.7%		
	8.1.10	Erie Drift Plain	25	0.1%			77	0.8%	143	0.3%	727	3.6%		+
	8.2.1	Southeastern Wisconsin Till Plains	129	0.6%			22	0.2%	233	0.6%	442	2.2%		
1	8.2.2	Huron/Erie Lake Plains	103	0.5%			177	1.8%	308	0.7%	133	0.7%		1
	8.2.3	Central Corn Belt Plains	23	0.1%					16	0.0%	308	1.5%		1
	8.2.4	Eastern Corn Belt Plains	45	0.2%			15	0.1%	9	0.0%	510	2.5%		
	8.3.1	Northern Piedmont	55	0.3%	30	0.0%	14	0.1%	6	0.0%	208	1.0%	2	0.0%
	8.3.2	Interior River Valleys and Hills	14	0.1%	34	0.1%	6	0.1%			485	2.4%		1
	8.3.3	Interior Plateau	91	0.4%	514	0.9%	21	0.2%			803	3.9%		
1	8.3.4	Piedmont	353	1.7%	13499	22.4%	7	0.1%			781	3.8%		
1	8.3.5	Southeastern Plains	1	0.0%	20564	34.1%	5	0.0%			929	4.5%		
1	8.3.6	Mississippi Valley Loess Plains			1302	2.2%					217	1.1%		
	8.3.7	South Central Plains			12048	20.0%					120	0.6%		
1	8.3.8	East Central Texas Plains			120	0.2%					4	0.0%		
	8.4.1	Ridge and Valley	1155	5.6%	1343	2.2%	81	0.8%	17	0.0%	1179	5.8%	1	0.0%
	8.4.2	Central Appalachians	198	1.0%	4	0.0%	77	0.8%	19	0.0%	1189	5.8%		
1	8.4.3	Western Allegheny Plateau	340	1.7%	98	0.2%	398	4.0%	90	0.2%	2285	11.2%		
	8.4.4	Blue Ridge	1531	7.5%	217	0.4%	1	0.0%			334	1.6%		1

			easter pi Median I Assoc I N/S	<b>n white</b> i <b>ne</b> N=8, S=6 N-↑, S-↓ = 0.59	Iobloll Media Assor N/S =	<b>y pine</b> n S=7 c S-↓ = 0.32	bigtooth Mediar Assoc N/S =	<b>1 aspen</b> n S=6 c S-↓ = 0.57	qual asp Media Assoc N/S	<b>cing</b> p <b>en</b> n N=7 c N-U = 0.6	black Mediar Asso N/S :	<b>cherry</b> n N=11 c N-U = 0.33	Dougla Median N= Assoc N- N/S =	<b>s-fir</b> =3, S=1 ↑, S-↓ 0.65
L2	CODE	US L3NAME	count	%	count	%	count	%	count	%	count	%	count	%
	8.4.5	Ozark Highlands			20	0.0%					392	1.9%		
1	8.4.6	Boston Mountains			48	0.1%					80	0.4%	-	
1	8.4.7	Arkansas Valley			170	0.3%					62	0.3%	-	
1	8.4.8	Ouachita Mountains			666	1.1%					83	0.4%		
1	8.4.9	Southwestern Appalachians	112	0.5%	1288	2.1%					211	1.0%		
	8.5.1	Middle Atlantic Coastal Plain			6065	10.0%	1	0.0%			192	0.9%		
1	8.5.2	Mississippi Alluvial Plain			87	0.1%					29	0.1%		
1	8.5.3	Southern Coastal Plain			1960	3.2%					64	0.3%		
1	8.5.4	Atlantic Coastal Pine Barrens	93	0.5%	2	0.0%	2	0.0%			26	0.1%		
	9.2.1	Northern Glaciated Plains						<u> </u>	301	0.7%				
	9.2.2	Lake Agassiz Plain				1			1548	3.7%	2	0.0%		
	9.2.3	Western Corn Belt Plains				1	1	0.0%	71	0.2%	180	0.9%		
	9.2.4	Central Irregular Plains		ll		1					159	0.8%		
	9.3.1	Northwestern Glaciated Plains			Í		1	1	9	0.0%				<u> </u>
	9.3.3	Northwestern Great Plains		ļ		1			24	0.1%	1	0.0%	190	0.5%
	9.3.4	Nebraska Sand Hills												
	9.4.1	High Plains				+							1	
	9.4.2	Central Great Plains		P									-	
	9.4.3	Southwestern Tablelands											-	
	9.4.4	Flint Hills		ľ		-								
	9.4.5	Cross Timbers				1					2	0.0%		
	9.4.6	Edwards Plateau											-	

			easter pi Median Assoc N/S	n white ine N=8, S=6 N-↑, S-↓ = 0.59	lobloll Media Assoc N/S =	<b>y pine</b> n S=7 c S-↓ ÷ 0.32	bigtooth Mediar Assoc N/S =	n <b>aspen</b> n S=6 c S-↓ ≤ 0.57	quak asp Mediar Assoc N/S =	<b>king</b> en n N=7 c N-U = 0.6	black Media Asso N/S :	<b>cherry</b> n N=11 c N-U = 0.33	Dougla Median N= Assoc N- N/S =	l <b>s-fir</b> =3, S=1 ↑, S-↓ 0.65
L2	CODE	US_L3NAME	count	%	count	%	count	%	count	%	count	%	count	%
	9.4.7	Texas Blackland Prairies			54	0.1%								
	9.5.1	Western Gulf Coastal Plain			237	0.4%					3	0.0%		
	9.6.1	Southern Texas Plains												
	10.1.2	Columbia Plateau							29	0.1%			102	0.3%
	10.1.3	Northern Basin and Range							80	0.2%			102	0.3%
	10.1.4	Wyoming Basin											25	0.1%
	10.1.5	Central Basin and Range							44	0.1%			21	0.1%
	10.1.6	Colorado Plateaus							187	0.4%			314	0.8%
1	10.1.7	Arizona/New Mexico Plateau											14	0.0%
1	10.1.8	Snake River Plain							24	0.1%				
	10.2.1	Mojave Basin and Range												
	10.2.2	Sonoran Basin and Range												
	10.2.10	Chihuahuan Deserts							-					
	11.1.1	Southern and Central California Chaparral and Oak Woodlands											173	0.4%
	11.1.2	Central California Valley												
	11.1.3	Southern California Mountains												
	12.1.1	Madrean Archipelago											20	0.1%
	13.1.1	Arizona/New Mexico Mountains							144	0.3%			486	1.2%
	15.4.1	Southern Florida Coastal Plain												
		Total Tree Cou			60374		10041		41748		20446		39364	

NA	NA 12		scar Media Asso N/S	<b>let oak</b> an N=10 bc N-U = 0.37	northe oa Mea N=10 Assoc N N/S =	ern pin ak dian ),S=5 √-↑, S-↓ = 0.41	southe oa Mediai Assoc N/S =	e <b>rn red</b> ak n N=9 c N-U = 0.36	bur Media Assoc N/S =	<b>oak</b> n N=9 c N-↓ : 0.59	wate Median M Assoc N N/S	<b>r oak</b> N=8, S=7 N-U, S-↓ = 0.26	<b>chestnu</b> Median Assoc N/S =	<b>it oak</b> N=9 N-U 0.45
L2	CODE	US_L3NAME	count	%	count	%	count	%	count	%	count	%	count	%
	5.2.1	Northern Lakes and Forests			1942	53.7%			2075	28.9%				
1	5.2.2	Northern Minnesota Wetlands			1	0.0%			128	1.8%				
	5.3.1	Northeastern Highlands	104	1.1%					2	0.0%			349	1.7%
1	5.3.3	North Central Appalachians	228	2.5%									1067	5.2%
	6.2.3	Northern Rockies	1											
ĺ	6.2.4	Canadian Rockies												
1	6.2.5	North Cascades												
1	6.2.7	Cascades												
Î.	6.2.8	Eastern Cascades Slopes and Foothills												
1	6.2.9	Blue Mountains												
1	6.2.10	Middle Rockies							128	1.8%				
1	6.2.11	Klamath Mountains												
1	6.2.12	Sierra Nevada												
1	6.2.13	Wasatch and Uinta Mountains												
1	6.2.14	Southern Rockies												
1	6.2.15	Idaho Batholith												
	7.1.7	Puget Lowland												
1	7.1.8	Coast Range												
	7.1.9	Willamette Valley												
	8.1.1	Eastern Great Lakes Lowlands			1	0.0%			31	0.4%			3	0.0%

NA_ NA_L3 L2 <u>CODE</u>		scarl Media Asso N/S	<b>et oak</b> in N=10 ic N-U = 0.37	northe oa Mec N=10 Assoc N N/S =	rn pin ak Jian ,S=5 I-↑, S-↓ = 0.41	southe oa Media Assoc N/S =	ern red ak n N=9 c N-U = 0.36	bur Media Assoc N/S =	<b>oak</b> n N=9 c N-↓ = 0.59	wate Median I Assoc N N/S	<b>r oak</b> N=8, S=7 N-U, S-↓ = 0.26	<b>chestnu</b> Median Assoc N/S =	<b>ut oak</b> ì N=9 N-U 0.45	
L2	CODE	US L3NAME	count	%	count	%	count	%	count	%	count	%	count	%
	8.1.3	Northern Allegheny Plateau	20	0.2%					3	0.0%			216	1.0%
	8.1.4	North Central Hardwood Forests			1158	32.0%			1503	20.9%				
	8.1.5	Driftless Area			220	6.1%			713	9.9%				
	8.1.6	Southern Michigan/Northern Indiana Drift Plains	6	0.1%	124	3.4%			41	0.6%				
	8.1.7	Northeastern Coastal Zone	567	6.2%					8	0.1%			174	0.8%
	8.1.8	Acadian Plains and Hills												
	8.1.10	Erie Drift Plain	4	0.0%					8	0.1%			5	0.0%
	8.2.1	Southeastern Wisconsin Till Plains			65	1.8%			129	1.8%				
	8.2.2	Huron/Erie Lake Plains	3	0.0%	65	1.8%			42	0.6%				
	8.2.3	Central Corn Belt Plains			2	0.1%			38	0.5%				
	8.2.4	Eastern Corn Belt Plains	4	0.0%					23	0.3%			4	0.0%
	8.3.1	Northern Piedmont	56	0.6%			75	1.0%					331	1.6%
	8.3.2	Interior River Valleys and Hills	64	0.7%			50	0.7%	43	0.6%			80	0.4%
	8.3.3	Interior Plateau	476	5.2%			404	5.4%	8	0.1%	59	0.5%	1010	4.9%
	8.3.4	Piedmont	786	8.6%			1431	19.1%			1374	11.1%	1180	5.7%
	8.3.5	Southeastern Plains	263	2.9%			1906	25.5%			5531	44.8%	231	1.1%
	8.3.6	Mississippi Valley Loess Plains	8	0.1%			217	2.9%	1	0.0%	454	3.7%		
	8.3.7	South Central Plains	1	0.0%			1292	17.3%			2058	16.7%		
	8.3.8	East Central Texas Plains					152	2.0%	1	0.0%	145	1.2%		
	8.4.1	Ridge and Valley	1507	16.4%	1	0.0%	320	4.3%	1	0.0%	123	1.0%	7106	34.3%
	8.4.2	Central Appalachians	745	8.1%			31	0.4%					2161	10.4%
	8.4.3	Western Allegheny Plateau	479	5.2%			11	0.1%					1127	5.4%
			scarl Media Asso N/S	l <b>et oak</b> in N=10 oc N-U = 0.37	northe oa Mec N=10 Assoc N N/S =	rn pin ik Jian ,S=5 I-↑, S-↓ = 0.41	southe oa Media Assoc N/S =	e <b>rn red</b> ak n N=9 c N-U = 0.36	bur Media Assoc N/S =	<b>oak</b> n N=9 c N-↓ = 0.59	wate Median I Assoc N N/S	e <b>r oak</b> N=8, S=7 N-U, S-↓ = 0.26	<b>chestni</b> Mediar Assoc N/S =	u <b>t oak</b> i N=9 N-U 0.45
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	CODE	US L3NAME	count	%	count	%	count	%	count	%	count	%	count	%
1	8.4.4	Blue Ridge	1507	16.4%			225	3.0%			20	0.2%	4206	20.3%
	8.4.5	Ozark Highlands	1365	14.9%			494	6.6%	12	0.2%	1	0.0%		
	8.4.6	Boston Mountains			<u> </u>		70	0.9%	6	0.1%				
	8.4.7	Arkansas Valley			<u> </u>		126	1.7%	1	0.0%	109	0.9%		
	8.4.8	Ouachita Mountains					107	1.4%			62	0.5%		
	8.4.9	Southwestern Appalachians	502	5.5%			266	3.6%			72	0.6%	1358	6.6%
	8.5.1	Middle Atlantic Coastal Plain	75	0.8%			190	2.5%			991	8.0%	24	0.1%
	8.5.2	Mississippi Alluvial Plain	1	0.0%			58	0.8%			209	1.7%		
Ì	8.5.3	Southern Coastal Plain	1	0.0%			17	0.2%			1039	8.4%		
Î	8.5.4	Atlantic Coastal Pine Barrens	394	4.3%			21	0.3%					79	0.4%
	9.2.1	Northern Glaciated Plains							340	4.7%				
	9.2.2	Lake Agassiz Plain							527	7.3%				
	9.2.3	Western Corn Belt Plains			29	0.8%			486	6.8%				
1	9.2.4	Central Irregular Plains	1	0.0%	8	0.2%	1	0.0%	151	2.1%				
	9.3.1	Northwestern Glaciated Plains							212	3.0%				
	9.3.3	Northwestern Great Plains							421	5.9%				
	9.3.4	Nebraska Sand Hills							16	0.2%				
	9.4.1	High Plains							3	0.0%				
	9.4.2	Central Great Plains	1						35	0.5%				
1	9.4.3	Southwestern Tablelands	1											
	9.4.4	Flint Hills							43	0.6%				
	9.4.5	Cross Timbers							1	0.0%				

			scarl Media Asso N/S	<b>et oak</b> n N=10 oc N-U = 0.37	northe oa Mec N=10 Assoc N N/S =	<b>rn pin</b> I <b>k</b> Iian ,S=5 I-↑, S-↓ : 0.41	southe oa Mediai Assoc N/S =	ern red ak n N=9 c N-U = 0.36	bur Mediar Assoc N/S =	oak n N=9 : N-↓ 0.59	wate Median I Assoc N N/S	e <b>r oak</b> N=8, S=7 N-U, S-↓ = 0.26	<b>chestnu</b> Median Assoc N/S =	n <b>t oak</b> N=9 N-U 0.45
L2	CODE	US_L3NAME	count	%	count	%	count	%	count	%	count	%	count	%
	9.4.6	Edwards Plateau												
	9.4.7	Texas Blackland Prairies					1	0.0%			3	0.0%		
	9.5.1	Western Gulf Coastal Plain					14	0.2%			102	0.8%		
	9.6.1	Southern Texas Plains												
	10.1.2	Columbia Plateau												
	10.1.3	Northern Basin and Range												
	10.1.4	Wyoming Basin									-			
1	10.1.5	Central Basin and Range												
	10.1.6	Colorado Plateaus												
1	10.1.7	Arizona/New Mexico Plateau												
	10.1.8	Snake River Plain												
	10.2.1	Mojave Basin and Range												
	10.2.2	Sonoran Basin and Range												
	10.2.10	Chihuahuan Deserts												
	11.1.1	Southern and Central California Chaparral and Oak Woodlands												
	11.1.2	Central California Valley												
	11.1.3	Southern California Mountains												
	12.1.1	Madrean Archipelago												
	13.1.1	Arizona/New Mexico Mountains												
	15.4.1	Southern Florida Coastal Plain												
		Total Tree Count	9167		3616		7479		7180		12352		20711	

			north o Media Asso N/S	<b>ern red</b> ak n N=10 oc N-↑ = 0.42	black Mediar Assoc N/S =	<b>x oak</b> n N=11 c N-↑ = 0.13	black Median S= Assoc N N/S =	<b>locust</b> N=11, 11 I-↑, S-↓ 0.18	black Mec N=10 Assoc N N/S =	<b>willow</b> lian ∖,S=7 I-U,S-↓ ± 0.29	sass Media Asso N/S	a <b>fras</b> n N=11 nc N-↑ = 0.28	pondcy Median Assoc N/S =	p <b>ress</b> N=7 N-↑ 0.71
L2	CODE	US L3NAME	count	%	count	%	count	%	count	%	count	%	count	%
	5.2.1	Northern Lakes and Forests	5778	20.2%	690	3.7%	28	0.7%	28	1.4%	22	0.4%		
1	5.2.2	Northern Minnesota Wetlands	2	0.0%										
	5.3.1	Northeastern Highlands	2993	10.5%	152	0.8%	28	0.7%	3	0.1%	25	0.5%		
1	5.3.3	North Central Appalachians	1020	3.6%	158	0.9%	6	0.2%	3	0.1%	221	4.4%		1
	6.2.3	Northern Rockies												
1	6.2.4	Canadian Rockies												1
1	6.2.5	North Cascades												
1	6.2.7	Cascades												1
1	6.2.8	Eastern Cascades Slopes and Foothills												
1	6.2.9	Blue Mountains												
1	6.2.10	Middle Rockies												
1	6.2.11	Klamath Mountains												
1	6.2.12	Sierra Nevada												
1	6.2.13	Wasatch and Uinta Mountains												
1	6.2.14	Southern Rockies												
1	6.2.15	Idaho Batholith						_						
	7.1.7	Puget Lowland												
1	7.1.8	Coast Range												1
1	7.1.9	Willamette Valley												1
	8.1.1	Eastern Great Lakes Lowlands	223	0.8%	23	0.1%	26	0.7%	40	2.0%	2	0.0%		<u> </u>
1	8.1.3	Northern Allegheny Plateau	822	2.9%	111	0.6%	51	1.3%	28	1.4%	8	0.2%		

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NA	NA 12		north o Media Asso N/S	ern red oak an N=10 oc N-↑ = 0.42	black Mediar Assoc N/S =	x <b>oak</b> n N=11 c N-↑ = 0.13	black I Median S= Assoc N N/S =	l <b>ocust</b> N=11, 11 I-↑, S-↓ 0.18	black Med N=10 Assoc N N/S =	<b>willow</b> lian ,S=7 I-U,S-↓ : 0.29	sass Media Asso N/S	<b>safras</b> n N=11 bc N-↑ = 0.28	<b>pondcy</b> Median Assoc N/S =	<b>press</b> N=7 N-↑ 0.71
L2	CODE	US_L3NAME	count	%	count	%	count	%	count	%	count	%	count	%
	8.1.4	North Central Hardwood Forests	1459	5.1%	827	4.5%	118	3.1%	74	3.6%				
	8.1.5	Driftless Area	1474	5.2%	636	3.4%	110	2.9%	25	1.2%				
	8.1.6	Southern Michigan/Northern Indiana Drift Plains	630	2.2%	684	3.7%	108	2.8%	68	3.3%	412	8.3%		
	8.1.7	Northeastern Coastal Zone	1657	5.8%	794	4.3%	52	1.4%	6	0.3%	41	0.8%		+
1	8.1.8	Acadian Plains and Hills	846	3.0%	8	0.0%								+
1	8.1.10	Erie Drift Plain	180	0.6%	46	0.2%	62	1.6%	29	1.4%	49	1.0%		+
	8.2.1	Southeastern Wisconsin Till Plains	141	0.5%	48	0.3%	88	2.3%	71	3.5%				+
1	8.2.2	Huron/Erie Lake Plains	175	0.6%	22	0.1%	6	0.2%	18	0.9%	58	1.2%		
1	8.2.3	Central Corn Belt Plains	60	0.2%	81	0.4%	74	1.9%	20	1.0%	39	0.8%		
1	8.2.4	Eastern Corn Belt Plains	200	0.7%	84	0.5%	148	3.9%	44	2.1%	115	2.3%		
	8.3.1	Northern Piedmont	150	0.5%	122	0.7%	68	1.8%	12	0.6%	76	1.5%		+
	8.3.2	Interior River Valleys and Hills	461	1.6%	686	3.7%	149	3.9%	170	8.3%	496	10.0%		
	8.3.3	Interior Plateau	664	2.3%	823	4.4%	338	8.8%	34	1.7%	785	15.8%		
1	8.3.4	Piedmont	773	2.7%	635	3.4%	68	1.8%	61	3.0%	52	1.0%		+
1	8.3.5	Southeastern Plains	165	0.6%	350	1.9%	24	0.6%	274	13.4%	111	2.2%	487	14.1%
1	8.3.6	Mississippi Valley Loess Plains	39	0.1%	65	0.4%	36	0.9%	105	5.1%	88	1.8%		
1	8.3.7	South Central Plains	6	0.0%	60	0.3%	7	0.2%	114	5.6%	85	1.7%	4	0.1%
	8.3.8	East Central Texas Plains			4	0.0%	2	0.1%	16	0.8%	15	0.3%		+
	8.4.1	Ridge and Valley	2335	8.2%	1371	7.4%	548	14.3%	8	0.4%	527	10.6%		
	8.4.2	Central Appalachians	1114	3.9%	573	3.1%	397	10.4%	9	0.4%	381	7.7%		1
1	8.4.3	Western Allegheny Plateau	793	2.8%	843	4.5%	437	11.4%	20	1.0%	668	13.4%		1
[	8.4.4	Blue Ridge	1240	4.3%	501	2.7%	428	11.2%	7	0.3%	161	3.2%		

NA	NA 12		north o Media Asso N/S	<b>ern red</b> ak n N=10 oc N-↑ = 0.42	black Mediar Assoc N/S =	<b>coak</b> nN=11 cN-↑ =0.13	black I Median S= Assoc N N/S =	ocust N=11, 11 I-↑, S-↓ 0.18	black Mec N=10 Assoc N N/S =	<b>willow</b> lian ,S=7 I-U,S-↓ : 0.29	sass Media Asso N/S	<b>safras</b> n N=11 bc N-↑ = 0.28	<b>pondcy</b> Mediar Assoc N/S =	<b>press</b> N=7 N-↑ 0.71
L2	CODE	US L3NAME	count	%	count	%	count	%	count	%	count	%	count	%
	8.4.5	Ozark Highlands	1331	4.7%	6319	34.0%	20	0.5%	9	0.4%	269	5.4%		
1	8.4.6	Boston Mountains	631	2.2%	418	2.3%	51	1.3%			29	0.6%		
1	8.4.7	Arkansas Valley	147	0.5%	112	0.6%	4	0.1%	22	1.1%	4	0.1%		
	8.4.8	Ouachita Mountains	364	1.3%	153	0.8%	4	0.1%	0	0.0%	1	0.0%		
	8.4.9	Southwestern Appalachians	273	1.0%	442	2.4%	38	1.0%	5	0.2%	110	2.2%		
	8.5.1	Middle Atlantic Coastal Plain	18	0.1%	67	0.4%	24	0.6%	71	3.5%	45	0.9%	109	3.2%
	8.5.2	Mississippi Alluvial Plain	7	0.0%	11	0.1%	9	0.2%	412	20.1%	12	0.2%	37	1.1%
	8.5.3	Southern Coastal Plain			2	0.0%			34	1.7%			2281	65.9%
	8.5.4	Atlantic Coastal Pine Barrens	13	0.0%	236	1.3%	7	0.2%	10	0.5%	59	1.2%		
	9.2.1	Northern Glaciated Plains												
1	9.2.2	Lake Agassiz Plain	2	0.0%					1	0.0%				
1	9.2.3	Western Corn Belt Plains	168	0.6%	67	0.4%	53	1.4%	72	3.5%				
1	9.2.4	Central Irregular Plains	198	0.7%	293	1.6%	160	4.2%	72	3.5%	3	0.1%		
	9.3.1	Northwestern Glaciated Plains												
1	9.3.3	Northwestern Great Plains												
	9.3.4	Nebraska Sand Hills							2	0.1%				
	9.4.1	High Plains			İ				1	0.0%				
	9.4.2	Central Great Plains					42	1.1%	22	1.1%				
	9.4.3	Southwestern Tablelands					3	0.1%	0	0.0%				
	9.4.4	Flint Hills	3	0.0%					2	0.1%				
	9.4.5	Cross Timbers	2	0.0%	42	0.2%			7	0.3%	2	0.0%		
1	9.4.6	Edwards Plateau												

NA			north o Media Asso N/S	<b>ern red</b> bak nn N=10 bc N-↑ = 0.42	black Median Assoc N/S =	a <b>oak</b> 1 N=11 2 N-↑ ÷ 0.13	black I Median S= Assoc N N/S =	<b>ocust</b> N=11, 11 I-↑, S-↓ 0.18	black Med N=10 Assoc N N/S =	<b>villow</b> lian ,S=7 I-U,S-↓ 0.29	sass Media Asso N/S	a <b>fras</b> n N=11 c N-↑ = 0.28	pondcy Median Assoc N/S =	p <b>ress</b> N=7 N-↑ 0.71
L2	CODE	US_L3NAME	count	%	count	%	count	%	count	%	count	%	count	%
	9.4.7	Texas Blackland Prairies												
	9.5.1	Western Gulf Coastal Plain							20	1.0%				
	9.6.1	Southern Texas Plains												
	10.1.2	Columbia Plateau												
1	10.1.3	Northern Basin and Range												
1	10.1.4	Wyoming Basin											-	
1	10.1.5	Central Basin and Range												
	10.1.6	Colorado Plateaus												
	10.1.7	Arizona/New Mexico Plateau												
1	10.1.8	Snake River Plain												
	10.2.1	Mojave Basin and Range												
	10.2.2	Sonoran Basin and Range												
	10.2.10	Chihuahuan Deserts												
	11.1.1	Southern and Central California Chaparral and Oak Woodlands												
	11.1.2	Central California Valley												
	11.1.3	Southern California Mountains												
	12.1.1	Madrean Archipelago												
	13.1.1	Arizona/New Mexico Mountains												
	15.4.1	Southern Florida Coastal Plain											541	15.6%
		Total Tree Count	28557		18559		3822		2049		4971		3459	

			baldc Media Asso N/S	<b>ypress</b> an S=6 oc S-↓ = 0.54	Ame bassy Median N Assoc N N/S	rican wood ∿=9, S=5 ŀ-↑, S-↓ = 0.4	east hem Media Assoc N/S =	t <b>ern</b> Iock n N=8 c N-↓ = 0.78	wes hem Media Assoc N/S =	<b>tern</b> <b>lock</b> n N=3 c N-U = 0.34	Ameri Median N Assoc N/S	<b>can elm</b> N=11, S=6 N-↑, S-↓ = 0.25	slipper Mediar Assoc N/S =	<b>y elm</b> ⊢S=8 S-↓ 0.09
L2	CODE	US L3NAME	count	%	count	%	count	%	count	%	count	%	count	%
	5.2.1	Northern Lakes and Forests			5591	44.4%	3098	13.5%			1082	7.6%	26	0.6%
1	5.2.2	Northern Minnesota Wetlands			139	1.1%					97	0.7%		
	5.3.1	Northeastern Highlands			200	1.6%	7154	31.3%			162	1.1%	3	0.1%
ľ	5.3.3	North Central Appalachians			148	1.2%	1271	5.6%			19	0.1%	2	0.0%
	6.2.3	Northern Rockies							705	7.5%				
1	6.2.4	Canadian Rockies							1	0.0%				+
1	6.2.5	North Cascades							1747	18.6%				
1	6.2.7	Cascades							4379	46.5%				+
1	6.2.8	Eastern Cascades Slopes and Foothills							59	0.6%				
1	6.2.9	Blue Mountains												
ľ	6.2.10	Middle Rockies									2	0.0%		
1	6.2.11	Klamath Mountains							28	0.3%				
1	6.2.12	Sierra Nevada												
1	6.2.13	Wasatch and Uinta Mountains												1
1	6.2.14	Southern Rockies												+
1	6.2.15	Idaho Batholith												
	7.1.7	Puget Lowland							179	1.9%				
	7.1.8	Coast Range							2307	24.5%				
	7.1.9	Willamette Valley							10	0.1%				
	8.1.1	Eastern Great Lakes Lowlands			276	2.2%	923	4.0%			358	2.5%	13	0.3%
	8.1.3	Northern Allegheny Plateau			325	2.6%	2010	8.8%			80	0.6%	1	0.0%

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			baldc Media Asso N/S	<b>eypress</b> an S=6 oc S-↓ = 0.54	Amer bassy Median N Assoc N N/S :	rican wood √=9, S=5 I-↑, S-↓ = 0.4	east hem Media Assoc N/S =	t <b>ern</b> Iock n N=8 c N-↓ = 0.78	west hem Mediar Assoc N/S =	tern lock n N=3 n N-U 0.34	Americ Median N Assoc I N/S	<b>can elm</b> I=11, S=6 N-↑, S-↓ = 0.25	<b>slippery</b> Median Assoc N/S =	<b>/ elm</b> S=8 S-↓ 0.09
L2	CODE	US L3NAME	count	%	count	%	count	%	count	%	count	%	count	%
	8.1.4	North Central Hardwood Forests			1698	13.5%	548	2.4%			1033	7.3%	131	3.2%
1	8.1.5	Driftless Area			969	7.7%					1512	10.6%	473	11.6%
1	8.1.6	Southern Michigan/Northern Indiana Drift Plains			352	2.8%	38	0.2%			566	4.0%	56	1.4%
1	8.1.7	Northeastern Coastal Zone			44	0.3%	1195	5.2%			119	0.8%	1	0.0%
1	8.1.8	Acadian Plains and Hills			44	0.3%	2657	11.6%			53	0.4%		
1	8.1.10	Erie Drift Plain			107	0.9%	383	1.7%			234	1.6%	45	1.1%
	8.2.1	Southeastern Wisconsin Till Plains			288	2.3%	14	0.1%			396	2.8%	54	1.3%
ľ	8.2.2	Huron/Erie Lake Plains			183	1.5%	7	0.0%			305	2.1%	28	0.7%
ľ	8.2.3	Central Corn Belt Plains			40	0.3%					125	0.9%	39	1.0%
1	8.2.4	Eastern Corn Belt Plains			232	1.8%					474	3.3%	140	3.4%
	8.3.1	Northern Piedmont			13	0.1%	9	0.0%			78	0.5%	20	0.5%
i i	8.3.2	Interior River Valleys and Hills	7	0.2%	77	0.6%					907	6.4%	286	7.0%
1 I	8.3.3	Interior Plateau	2	0.1%	100	0.8%	1	0.0%			674	4.7%	437	10.7%
1 I	8.3.4	Piedmont	4	0.1%	10	0.1%	26	0.1%			273	1.9%	142	3.5%
1 I	8.3.5	Southeastern Plains	549	19.0%	23	0.2%	9	0.0%			371	2.6%	168	4.1%
1	8.3.6	Mississippi Valley Loess Plains	56	1.9%	1	0.0%					233	1.6%	127	3.1%
1	8.3.7	South Central Plains	458	15.8%	9	0.1%					287	2.0%	75	1.8%
İ.	8.3.8	East Central Texas Plains			4	0.0%					43	0.3%		
	8.4.1	Ridge and Valley			295	2.3%	966	4.2%			166	1.2%	141	3.4%
1	8.4.2	Central Appalachians			556	4.4%	908	4.0%			87	0.6%	92	2.3%
1	8.4.3	Western Allegheny Plateau			154	1.2%	347	1.5%			659	4.6%	498	12.2%
	8.4.4	Blue Ridge			209	1.7%	1018	4.5%			16	0.1%	13	0.3%

			baldc Media Asso N/S	<b>eypress</b> an S=6 oc S-↓ = 0.54	Amer bassy Median N Assoc N N/S	rican wood √=9, S=5 I-↑, S-↓ = 0.4	east hem Mediar Assoc N/S =	<b>ern</b> I <b>ock</b> n N=8 c N-↓ i 0.78	west hem Mediat Assoc N/S =	tern lock n N=3 N-U 0.34	Americ Median N Assoc I N/S	<b>can elm</b> I=11, S=6 N-↑, S-↓ = 0.25	<b>slippery</b> Median Assoc N/S =	<b>/ elm</b> S=8 S-↓ 0.09
L2	CODE	US_L3NAME	count	%	count	%	count	%	count	%	count	%	count	%
	8.4.5	Ozark Highlands			21	0.2%					687	4.8%	387	9.5%
1	8.4.6	Boston Mountains			20	0.2%					27	0.2%	35	0.9%
1	8.4.7	Arkansas Valley			<u> </u>						37	0.3%	16	0.4%
1	8.4.8	Ouachita Mountains			2	0.0%					20	0.1%	13	0.3%
1	8.4.9	Southwestern Appalachians			54	0.4%	282	1.2%			35	0.2%	21	0.5%
	8.5.1	Middle Atlantic Coastal Plain	344	11.9%	0	0.0%					107	0.8%	35	0.9%
1	8.5.2	Mississippi Alluvial Plain	525	18.2%	2	0.0%					449	3.2%	203	5.0%
1	8.5.3	Southern Coastal Plain	793	27.4%	1	0.0%					205	1.4%	14	0.3%
1	8.5.4	Atlantic Coastal Pine Barrens			0	0.0%							1	0.0%
	9.2.1	Northern Glaciated Plains			9	0.1%					18	0.1%		
1	9.2.2	Lake Agassiz Plain			102	0.8%					103	0.7%	1	0.0%
1	9.2.3	Western Corn Belt Plains			232	1.8%					654	4.6%	190	4.6%
İ	9.2.4	Central Irregular Plains			48	0.4%					1001	7.0%	126	3.1%
	9.3.1	Northwestern Glaciated Plains			4	0.0%					47	0.3%	2	0.0%
1	9.3.3	Northwestern Great Plains									63	0.4%		
1	9.3.4	Nebraska Sand Hills									1	0.0%		
	9.4.1	High Plains									7	0.0%		
	9.4.2	Central Great Plains									174	1.2%	14	0.3%
1	9.4.3	Southwestern Tablelands									7	0.0%		
1	9.4.4	Flint Hills			5	0.0%					103	0.7%	10	0.2%
1	9.4.5	Cross Timbers									32	0.2%	3	0.1%
[	9.4.6	Edwards Plateau												

			baldc Media Asso N/S	<b>ypress</b> an S=6 oc S-↓ = 0.54	Amer bassw Median N Assoc N N/S =	rican vood I=9, S=5 I-↑, S-↓ = 0.4	east hem Media Assoc N/S =	t <b>ern</b> Iock n N=8 c N-↓ = 0.78	west hem Mediat Assoc N/S =	tern lock n N=3 c N-U c 0.34	Americ Median N Assoc I N/S	<b>can elm</b> J=11, S=6 N-↑, S-↓ = 0.25	slippery Median Assoc N/S = 1	<b>/ elm</b> S=8 S-↓ 0.09
L2	CODE	US_L3NAME	count	%	count	%	count	%	count	%	count	%	count	%
	9.4.7	Texas Blackland Prairies									2	0.0%		
	9.5.1	Western Gulf Coastal Plain	16	0.6%							20	0.1%	5	0.1%
	9.6.1	Southern Texas Plains												
	10.1.2	Columbia Plateau												
	10.1.3	Northern Basin and Range									-			
	10.1.4	Wyoming Basin									-			
	10.1.5	Central Basin and Range												
	10.1.6	Colorado Plateaus									-			
	10.1.7	Arizona/New Mexico Plateau									-			
	10.1.8	Snake River Plain												
	10.2.1	Mojave Basin and Range												
	10.2.2	Sonoran Basin and Range												
	10.2.10	Chihuahuan Deserts									-			
	11.1.1	Southern and Central California Chaparral and Oak Woodlands												
	11.1.2	Central California Valley												
	11.1.3	Southern California Mountains												
	12.1.1	Madrean Archipelago												
	13.1.1	Arizona/New Mexico Mountains												
	15.4.1	Southern Florida Coastal Plain	138	4.8%										
		Total Tree Count	2892		12587		22864		9415		14210		4087	

1	Attachment 2B
2	
3	Species-specific Sample Distribution across Ecoregions
4	for Species with Statistically Significant Associations of Survival with N/S
5	from Horn et al 2018 Supplemental Information Dataset

## 6 **Key:**

- 7 NA\_L2 = North American Ecoregion, code for level II
- 8 NA\_L3 = North American Ecoregion, code for level III
- 9 US\_L3NAME = Name of Ecoregion at level III
- 10 See: <u>https://www.epa.gov/eco-research/ecoregions</u>
- 11 Median = Tree-specific median S and/or N deposition for the species samples
- 12 Assoc = U= unimodal,  $\uparrow$ =positive,  $\downarrow$ =negative
- 13 N/S = correlation coefficient for N and S deposition values for the species samples
- 14 Count = number of species' tree samples assessed in all plots in that ecoregion
- 15 % = percent of species' tree samples in that ecoregion

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			box Media Asso N/S :	<b>elder</b> n S = 6 bc S-↓ = 0.13	red m Median N Assoc N N/S =	n <b>aple</b> ↓=9, S=7 I-U, S-↓ 0.59	sugar Media Assoc N/S =	<b>maple</b> n S=8 c S-↓ 5 0.67	<b>yellow</b> Mediar Asso N/S =	<b>birch</b> n S = 5 c S-↓ 0.71	swee Median N Assoc I N/S :	<b>t birch</b> =10, S =13 N-U, S-↓ = 0.57	<b>paper I</b> Median N Assoc N- NS = (	<b>ɔirch</b> =7, S=4 ·U, S-↓ ).42
NA_ L2	NA_L3 CODE	US_L3NAME	count	%	count	%	count	%	count	%	count	%	count	%
5.2	5.2.1	Northern Lakes and Forests	97	1.3%	26666	22.0%	31512	42.2%	3912	24.0%			12403	50.0%
5.Z	5.2.2	Northern Minnesota Wetlands	61	0.8%	98	0.1%	77	0.1%	2	0.0%			657	2.6%
5.2	5.3.1	Northeastern Highlands	24	0.3%	15529	12.8%	12843	17.2%	7577	46.6%	1471	14.4%	6728	27.1%
5.5	5.3.3	North Central Appalachians			5908	4.9%	2186	2.9%	452	2.8%	1477	14.5%	122	0.5%
	6.2.3	Northern Rockies											219	0.9%
	6.2.4	Canadian Rockies											10	0.0%
	6.2.5	North Cascades											2	0.0%
	6.2.7	Cascades												
	6.2.8	Eastern Cascades Slopes and Foothills												
6.2	6.2.9	Blue Mountains											4	0.0%
0.2	6.2.10	Middle Rockies											28	0.1%
	6.2.11	Klamath Mountains												
	6.2.12	Sierra Nevada												
	6.2.13	Wasatch and Uinta Mountains												
	6.2.14	Southern Rockies												
	6.2.15	Idaho Batholith											12	0.0%
	7.1.7	Puget Lowland											24	0.1%
7.1	7.1.8	Coast Range												
	7.1.9	Willamette Valley												
	8.1.1	Eastern Great Lakes Lowlands	111	1.5%	1943	1.6%	1122	1.5%	240	1.5%	56	0.5%	80	0.3%
8.1	8.1.3	Northern Allegheny Plateau	10	0.1%	4347	3.6%	3421	4.6%	466	2.9%	626	6.1%	110	0.4%
	8.1.4	North Central Hardwood Forests	684	9.1%	4556	3.8%	3346	4.5%	488	3.0%			1249	5.0%

			box Media Asso N/S	<b>elder</b> In S = 6 Dc S-↓ = 0.13	red m Median N Assoc N N/S =	<b>iaple</b> I=9, S=7 I-U, S-↓ 0.59	sugar Mediar Assoc N/S =	<b>maple</b> n S=8 c S-↓ 0.67	<b>yellow</b> Mediar Assoc N/S =	<b>birch</b> n S = 5 c S-↓ 0.71	sweet Median N Assoc N N/S =	t <b>birch</b> =10, S =13 √-U, S-↓ = 0.57	<b>paper k</b> Median N= Assoc N- NS = 0	<b>birch</b> =7, S=4 U, S-↓ ).42
L2	NA_L3 CODE	US_L3NAME	count	%	count	%	count	%	count	%	count	%	count	%
	8.1.5	Driftless Area	1095	14.6%	750	0.6%	916	1.2%	32	0.2%			891	3.6%
1	8.1.6	Southern Michigan/Northern Indiana Drift Plains	142	1.9%	1923	1.6%	631	0.8%	42	0.3%			39	0.2%
1	8.1.7	Northeastern Coastal Zone	7	0.1%	5239	4.3%	510	0.7%	300	1.8%	1095	10.7%	181	0.7%
	8.1.8	Acadian Plains and Hills			5482	4.5%	1267	1.7%	1347	8.3%			1784	7.2%
	8.1.10	Erie Drift Plain	8	0.1%	2203	1.8%	984	1.3%	124	0.8%	10	0.1%		
	8.2.1	Southeastern Wisconsin Till Plains	404	5.4%	198	0.2%	229	0.3%	61	0.4%			72	0.3%
0.2	8.2.2	Huron/Erie Lake Plains	103	1.4%	1283	1.1%	117	0.2%	10	0.1%			157	0.6%
0.2	8.2.3	Central Corn Belt Plains	103	1.4%	17	0.0%	63	0.1%						
	8.2.4	Eastern Corn Belt Plains	251	3.3%	462	0.4%	1233	1.6%	2	0.0%				
	8.3.1	Northern Piedmont	92	1.2%	759	0.6%	99	0.1%	4	0.0%	144	1.4%		
	8.3.2	Interior River Valleys and Hills	370	4.9%	588	0.5%	1377	1.8%						
	8.3.3	Interior Plateau	600	8.0%	1367	1.1%	4029	5.4%						
83	8.3.4	Piedmont	195	2.6%	4099	3.4%	14	0.0%	2	0.0%	29	0.3%		
0.5	8.3.5	Southeastern Plains	207	2.8%	5825	4.8%	47	0.1%						
	8.3.6	Mississippi Valley Loess Plains	252	3.4%	261	0.2%	74	0.1%						
	8.3.7	South Central Plains	120	1.6%	1001	0.8%	2	0.0%						
	8.3.8	East Central Texas Plains	7	0.1%	5	0.0%								
	8.4.1	Ridge and Valley	131	1.7%	6002	4.9%	1707	2.3%	201	1.2%	2133	20.9%	20	0.1%
	8.4.2	Central Appalachians	25	0.3%	5895	4.9%	2534	3.4%	594	3.7%	1347	13.2%		
8.4	8.4.3	Western Allegheny Plateau	249	3.3%	4725	3.9%	2779	3.7%	17	0.1%	265	2.6%		
	8.4.4	Blue Ridge	23	0.3%	4545	3.7%	397	0.5%	395	2.4%	1524	14.9%		
	8.4.5	Ozark Highlands	121	1.6%	219	0.2%	531	0.7%						

			box Media Asso N/S	a <b>elder</b> an S = 6 bc S-↓ = 0.13	red m Median N Assoc N N/S =	<b>iaple</b> I=9, S=7 I-U, S-↓ 0.59	sugar Mediar Assoc N/S =	<b>maple</b> n S=8 c S-↓ 0.67	yellow Median Assoc N/S =	<b>birch</b> S = 5 S-↓ 0.71	swee Median N Assoc N N/S =	t <b>birch</b> =10, S =13 ∿-U, S-↓ = 0.57	<b>paper k</b> Median N= Assoc N- NS = (	<b>&gt;irch</b> =7, S=4 U, S-↓ ).42
NA_ L2	NA_L3 CODE	US_L3NAME	count	%	count	%	count	%	count	%	count	%	count	%
	8.4.6	Boston Mountains			223	0.2%	26	0.0%						
	8.4.7	Arkansas Valley	36	0.5%	68	0.1%	1	0.0%						
	8.4.8	Ouachita Mountains	5	0.1%	197	0.2%								
	8.4.9	Southwestern Appalachians	27	0.4%	1760	1.5%	527	0.7%	2	0.0%	30	0.3%		
	8.5.1	Middle Atlantic Coastal Plain	23	0.3%	4009	3.3%								
	8.5.2	Mississippi Alluvial Plain	524	7.0%	716	0.6%	8	0.0%						
8.5	8.5.3	Southern Coastal Plain	4	0.1%	1984	1.6%								
	8.5.4	Atlantic Coastal Pine Barrens	1	0.0%	325	0.3%					8	0.1%		
	9.2.1	Northern Glaciated Plains	157	2.1%									6	0.0%
	9.2.2	Lake Agassiz Plain	221	2.9%	3	0.0%	39						8	0.0%
9.2	9.2.3	Western Corn Belt Plains	692	9.2%	30	0.0%	78	0.1%	3	0.0%			9	0.0%
	9.2.4	Central Irregular Plains	202	2.7%	2	0.0%	34	0.0%						
	9.3.1	Northwestern Glaciated Plains	21	0.3%										
9.3	9.3.3	Northwestern Great Plains	14	0.2%										
	9.3.4	Nebraska Sand Hills	1	0.0%										
	9.4.1	High Plains	4	0.1%										
	9.4.2	Central Great Plains	62	0.8%										
	9.4.3	Southwestern Tablelands	4	0.1%										
9.4	9.4.4	Flint Hills	8	0.1%										
	9.4.5	Cross Timbers	4	0.1%										
	9.4.6	Edwards Plateau												
	9.4.7	Texas Blackland Prairies												

			box Media Asso N/S	<b>elder</b> In S = 6 Dc S-↓ = 0.13	red m Median N Assoc N N/S =	<b>iaple</b> I=9, S=7 I-U, S-↓ 0.59	sugar Mediai Assoc N/S =	<b>maple</b> n S=8 c S-↓ 0.67	yellow Median Assoc N/S =	<b>birch</b> S = 5 S-↓ 0.71	sweet Median N Assoc N N/S =	t <b>birch</b> =10, S =13 √-U, S-↓ = 0.57	<b>paper b</b> Median N= Assoc N- NS = 0	<b>&gt;irch</b> =7, S=4 U, S-↓ ).42
NA_ L2	NA_L3 CODE	US_L3NAME	count	%	count	%	count	%	count	%	count	%	count	%
9.5	9.5.1	Western Gulf Coastal Plain			11	0.0%								
9.6	9.6.1	Southern Texas Plains												
	10.1.2	Columbia Plateau												
	10.1.3	Northern Basin and Range												
	10.1.4	Wyoming Basin												
10.1	10.1.5	Central Basin and Range												
	10.1.6	Colorado Plateaus												
	10.1.7	Arizona/New Mexico Plateau												
	10.1.8	Snake River Plain												
10.2	10.2.1	Mojave Basin and Range												
	10.2.2	Sonoran Basin and Range												
	10.2.10	Chihuahuan Deserts												
	11.1.1	Southern and Central California Chaparral and Oak Woodlands												
11.1	11.1.2	Central California Valley	10	0.1%										
	11.1.3	Southern California Mountains												
12.1	12.1.1	Madrean Archipelago												
13.1	13.1.1	Arizona/New Mexico Mountains	1	0.0%										
15.4	15.4.1	Southern Florida Coastal Plain			65	0.1%								
		Total Tree Count	7513		121288		74760		16273		10215		24815	

NA			Ame horr Medi Asse N/S	erican 1beam an S=7 ⊃c S-↓ = 0.26	mock hick Mediar Assoc N/S =	ernut xory n N=10 c N-↓ = 0.15	<b>pignut I</b> Median Assoc N/S =	nickory n S=10 c S-↓ = 0.4	hackl Media Assoc N/S =	<b>berry</b> n S=7 c S-↓ ÷ 0.18	America Median Assoc N N/S	an beech N=8, S=7 N-U, S-↓ = 0.76	white Median Assoc N/S =	<b>ash</b> N=10 N-U 0.53
L2	CODE	US_L3NAME	count	%	count	%	count	%	count	%	count	%	count	%
5.2	5.2.1	Northern Lakes and Forests	14	0.4%							1565	6.4%	1415	7.0%
1	5.2.2	Northern Minnesota Wetlands							1	0.0%				
5.3	5.3.1	Northeastern Highlands	14	0.4%	30	0.3%	171	1.4%			9630	39.5%	2787	13.8%
	5.3.3	North Central Appalachians	51	1.6%	13	0.1%	97	0.8%			1747	7.2%	541	2.7%
6.2	6.2.3	Northern Rockies												
	6.2.4	Canadian Rockies												+
1	6.2.5	North Cascades												
	6.2.7	Cascades			<u> </u>									
1	6.2.8	Eastern Cascades Slopes and Foothills												
	6.2.9	Blue Mountains			<u> </u>									
1	6.2.10	Middle Rockies							1	0.0%				
	6.2.11	Klamath Mountains												
	6.2.12	Sierra Nevada			<u> </u>									
	6.2.13	Wasatch and Uinta Mountains			<u> </u>									
1	6.2.14	Southern Rockies												
	6.2.15	Idaho Batholith			<u> </u>									
7.1	7.1.7	Puget Lowland												
	7.1.8	Coast Range												
	7.1.9	Willamette Valley												+
8.1	8.1.1	Eastern Great Lakes Lowlands	9	0.3%			20	0.2%	4	0.1%	324	1.3%	757	3.7%
1	8.1.3	Northern Allegheny Plateau	35	1.1%	9	0.1%	97	0.8%			1415	5.8%	1958	9.7%
	8.1.4	North Central Hardwood Forests	6	0.2%					49	0.9%	198	0.8%	657	3.2%

NA	NA 13		Ame horr Media Asso N/S	erican Ibeam an S=7 oc S-↓ = 0.26	mock hick Median Assoc N/S =	ernut ory n N=10 c N-↓ 0.15	<b>pignut ł</b> Median Assoc N/S =	nickory n S=10 c S-↓ = 0.4	hackl Mediar Assoc N/S =	<b>berry</b> n S=7 c S-↓ ∈ 0.18	America Median N Assoc N N/S =	I <b>n beech</b> N=8, S=7 I-U, S-↓ = 0.76	white Median Assoc N/S =	<b>ash</b> N=10 N-U 0.53
L2	CODE	US_L3NAME	count	%	count	%	count	%	count	%	count	%	count	%
	8.1.5	Driftless Area	2	0.1%					251	4.5%			399	2.0%
	8.1.6	Southern Michigan/Northern Indiana Drift Plains	12	0.4%	20	0.2%	138	1.1%	39	0.7%	177	0.7%	370	1.8%
1	8.1.7	Northeastern Coastal Zone	6	0.2%	60	0.5%	253	2.1%	5	0.1%	375	1.5%	503	2.5%
	8.1.8	Acadian Plains and Hills	0	0.0%							1672	6.9%	788	3.9%
1	8.1.10	Erie Drift Plain	14	0.4%	5	0.0%	23	0.2%	4	0.1%	358	1.5%	558	2.8%
8.2	8.2.1	Southeastern Wisconsin Till Plains	0	0.0%					26	0.5%	47	0.2%	196	1.0%
	8.2.2	Huron/Erie Lake Plains	2	0.1%	3	0.0%	13	0.1%	14	0.3%	33	0.1%	135	0.7%
	8.2.3	Central Corn Belt Plains	3	0.1%	8	0.1%	3	0.0%	89	1.6%			55	0.3%
1	8.2.4	Eastern Corn Belt Plains	12	0.4%	47	0.4%	115	0.9%	309	5.6%	135	0.6%	885	4.4%
8.3	8.3.1	Northern Piedmont	13	0.4%	197	1.7%	230	1.9%	32	0.6%	97	0.4%	321	1.6%
	8.3.2	Interior River Valleys and Hills	17	0.5%	296	2.6%	673	5.5%	637	11.4%	155	0.6%	591	2.9%
1	8.3.3	Interior Plateau	95	3.0%	782	6.9%	2090	17.2%	1210	21.7%	914	3.7%	1695	8.4%
1	8.3.4	Piedmont	406	12.6%	1261	11.1%	1301	10.7%	80	1.4%	626	2.6%	347	1.7%
1	8.3.5	Southeastern Plains	879	27.3%	1163	10.2%	982	8.1%	63	1.1%	758	3.1%	129	0.6%
1	8.3.6	Mississippi Valley Loess Plains	253	7.9%	211	1.9%	263	2.2%	10	0.2%	134	0.5%	96	0.5%
	8.3.7	South Central Plains	708	22.0%	497	4.4%	68	0.6%	9	0.2%	207	0.8%	163	0.8%
	8.3.8	East Central Texas Plains	6	0.2%	29	0.3%	6	0.0%	2	0.0%			37	0.2%
8.4	8.4.1	Ridge and Valley	23	0.7%	1025	9.0%	1455	11.9%	151	2.7%	508	2.1%	1076	5.3%
1	8.4.2	Central Appalachians	53	1.6%	580	5.1%	786	6.5%	6	0.1%	1724	7.1%	511	2.5%
	8.4.3	Western Allegheny Plateau	44	1.4%	815	7.2%	952	7.8%	79	1.4%	829	3.4%	1367	6.7%
1	8.4.4	Blue Ridge	23	0.7%	511	4.5%	804	6.6%	5	0.1%	353	1.4%	395	1.9%
	8.4.5	Ozark Highlands	9	0.3%	1530	13.4%	664	5.4%	294	5.3%	1	0.0%	649	3.2%

ΝA	NA 13		Ame horr Media Asso N/S	erican Ibeam an S=7 oc S-↓ = 0.26	mock hick Median Assoc N/S =	ernut ory N=10 N-↓ 0.15	<b>pignut I</b> Median Assoc N/S =	nickory n S=10 c S-↓ = 0.4	hackl Mediar Assoc N/S =	<b>perry</b> n S=7 c S-↓ : 0.18	America Median Assoc N N/S	an beech N=8, S=7 N-U, S-↓ = 0.76	white Median Assoc N/S =	<b>ash</b> N=10 N-U 0.53
L2	CODE	US_L3NAME	count	%	count	%	count	%	count	%	count	%	count	%
	8.4.6	Boston Mountains	5	0.2%	619	5.4%	24	0.2%	24	0.4%	65	0.3%	102	0.5%
	8.4.7	Arkansas Valley	14	0.4%	288	2.5%	1	0.0%	40	0.7%			79	0.4%
	8.4.8	Ouachita Mountains	37	1.2%	565	5.0%	2	0.0%	6	0.1%	9	0.0%	29	0.1%
1	8.4.9	Southwestern Appalachians	38	1.2%	536	4.7%	754	6.2%	35	0.6%	199	0.8%	240	1.2%
8.5	8.5.1	Middle Atlantic Coastal Plain	170	5.3%	110	1.0%	58	0.5%	26	0.5%	109	0.4%	41	0.2%
	8.5.2	Mississippi Alluvial Plain	39	1.2%	85	0.7%	29	0.2%	71	1.3%	11	0.0%	9	0.0%
	8.5.3	Southern Coastal Plain	186	5.8%	14	0.1%	96	0.8%	10	0.2%	3	0.0%	16	0.1%
	8.5.4	Atlantic Coastal Pine Barrens	3	0.1%	4	0.0%	6	0.0%	0	0.0%	19	0.1%	7	0.0%
9.2	9.2.1	Northern Glaciated Plains							17	0.3%				
	9.2.2	Lake Agassiz Plain												
	9.2.3	Western Corn Belt Plains			2	0.0%	2	0.0%	651	11.7%			104	0.5%
1	9.2.4	Central Irregular Plains			69	0.6%	7	0.1%	855	15.4%			243	1.2%
9.3	9.3.1	Northwestern Glaciated Plains							16	0.3%				
	9.3.3	Northwestern Great Plains							3	0.1%				
	9.3.4	Nebraska Sand Hills							7	0.1%				
9.4	9.4.1	High Plains			İ				2	0.0%				
	9.4.2	Central Great Plains							260	4.7%			2	0.0%
	9.4.3	Southwestern Tablelands							11	0.2%				
	9.4.4	Flint Hills							151	2.7%			2	0.0%
	9.4.5	Cross Timbers			5	0.0%	1	0.0%	10	0.2%			8	0.0%
	9.4.6	Edwards Plateau												1
	9.4.7	Texas Blackland Prairies			1	0.0%					<u> </u>		3	0.0%

NA			Ame horn Media Asso N/S	erican Ibeam an S=7 oc S-↓ = 0.26	mock hick Median Assoc N/S =	ernut cory n N=10 c N-↓ = 0.15	<b>pignut I</b> Mediar Assoc N/S	hickory n S=10 c S-↓ = 0.4	hackt Mediar Assoc N/S =	<b>berry</b> n S=7 ≳ S-↓ ⊴ 0.18	America Median I Assoc N N/S	<b>In beech</b> N=8, S=7 I-U, S-↓ = 0.76	white a Median I Assoc I N/S = (	<b>ash</b> N=10 N-U 0.53
L2	CODE	US_L3NAME	count	%	count	%	count	%	count	%	count	%	count	%
9.5	9.5.1	Western Gulf Coastal Plain	13	0.4%	2	0.0%	1	0.0%						
9.6	9.6.1	Southern Texas Plains												
10.1	10.1.2	Columbia Plateau												
1	10.1.3	Northern Basin and Range												
	10.1.4	Wyoming Basin												
	10.1.5	Central Basin and Range											-	
	10.1.6	Colorado Plateaus												
	10.1.7	Arizona/New Mexico Plateau												
	10.1.8	Snake River Plain												
10.2	10.2.1	Mojave Basin and Range												
	10.2.2	Sonoran Basin and Range												
	10.2.10	Chihuahuan Deserts									-		-	
11.1	11.1.1	Southern and Central California Chaparral and Oak Woodlands												
	11.1.2	Central California Valley												
1	11.1.3	Southern California Mountains												
12.1	12.1.1	Madrean Archipelago												
13.1	13.1.1	Arizona/New Mexico Mountains												
15.4	15.4.1	Southern Florida Coastal Plain												
		Total Tree Count	3214		11392		12185		5565		24397		20266	

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			gree Me N=1 Assoc N/S	<b>en ash</b> edian 0,S=6 N-U, S-↓ = 0.45	black v Mediar Assoc N/S =	walnut n N=12 c N-U = 0.13	Utah ju Media Assoc N/S =	uniper n N=3 c N-U : 0.72	east redco Mec N=11 Assoc N N/S Col	t <b>ern</b> edar lian ,S=7 I-U,S-↓ r r= 0.3	swee Median I Assoc N N/S	e <b>tgum</b> N=9, S=7 N-U, S-↓ = 0.37	<b>yellow p</b> Median Assoc N/S =	o <b>plar</b> S=11 S-↓ 0.4
L2	CODE	US_L3NAME	count	%	count	%	count	%	count	%	count	%	count	%
5.2	5.2.1	Northern Lakes and Forests	1941	10.3%	4	0.1%			2	0.0%				
1	5.2.2	Northern Minnesota Wetlands	216	1.1%										
5.3	5.3.1	Northeastern Highlands	45	0.2%	6	0.1%			28	0.2%			112	0.4%
1	5.3.3	North Central Appalachians			1	0.0%			4	0.0%			57	0.2%
6.2	6.2.3	Northern Rockies	1											
İ	6.2.4	Canadian Rockies												
1	6.2.5	North Cascades												
1	6.2.7	Cascades												
ĺ	6.2.8	Eastern Cascades Slopes and Foothills												
1	6.2.9	Blue Mountains												
1	6.2.10	Middle Rockies	25	0.1%			60	0.3%						
1	6.2.11	Klamath Mountains												
1	6.2.12	Sierra Nevada												
1	6.2.13	Wasatch and Uinta Mountains					1089	5.8%						
ĺ	6.2.14	Southern Rockies					219	1.2%						
1	6.2.15	Idaho Batholith												
7.1	7.1.7	Puget Lowland												
1	7.1.8	Coast Range												
1	7.1.9	Willamette Valley												
8.1	8.1.1	Eastern Great Lakes Lowlands	546	2.9%	40	0.6%			48	0.3%			9	0.0%

			gree Me N=1 Assoc I N/S	n ash dian 0,S=6 N-U, S-↓ = 0.45	black Mediar Assoc N/S =	walnut n N=12 c N-U = 0.13	Utah ju Mediar Assoc N/S =	<b>uniper</b> n N=3 e N-U e 0.72	east redc Mec N=11 Assoc N N/S Co	t <b>ern</b> edar Jian ,S=7 I-U,S-↓ r r= 0.3	swee Median N Assoc N N/S =	t <b>gum</b> N=9, S=7 N-U, S-↓ = 0.37	yellow p Median Assoc N/S =	<b>oplar</b> S=11 S-↓ : 0.4
L2	NA_L3	US L3NAME	count	%	count	%	count	%	count	%	count	%	count	%
	8.1.3	Northern Allegheny Plateau	99	0.5%	40	0.6%			21	0.1%			7	0.0%
	8.1.4	North Central Hardwood Forests	1561	8.3%	13	0.2%			123	0.7%				
1	8.1.5	Driftless Area	217	1.2%	448	6.8%			313	1.8%				
	8.1.6	Southern Michigan/Northern Indiana Drift Plains	1128	6.0%	145	2.2%			27	0.2%			71	0.3%
	8.1.7	Northeastern Coastal Zone	62	0.3%	23	0.3%			172	1.0%	2	0.0%	58	0.2%
	8.1.8	Acadian Plains and Hills	31	0.2%										
1	8.1.10	Erie Drift Plain	162	0.9%	63	1.0%					1	0.0%	184	0.7%
8.2	8.2.1	Southeastern Wisconsin Till Plains	735	3.9%	88	1.3%			102	0.6%				
	8.2.2	Huron/Erie Lake Plains	813	4.3%	42	0.6%			1	0.0%			4	0.0%
	8.2.3	Central Corn Belt Plains	126	0.7%	148	2.2%			8	0.0%			6	0.0%
	8.2.4	Eastern Corn Belt Plains	359	1.9%	469	7.1%			161	0.9%	84	0.2%	206	0.7%
8.3	8.3.1	Northern Piedmont	61	0.3%	171	2.6%			277	1.6%	63	0.2%	804	2.9%
	8.3.2	Interior River Valleys and Hills	666	3.5%	514	7.8%			738	4.3%	569	1.5%	436	1.6%
	8.3.3	Interior Plateau	904	4.8%	931	14.1%			3994	23.2%	1003	2.7%	2609	9.4%
	8.3.4	Piedmont	621	3.3%	192	2.9%			1329	7.7%	6822	18.3%	5776	20.9%
	8.3.5	Southeastern Plains	1302	6.9%	66	1.0%			873	5.1%	11773	31.6%	4012	14.5%
	8.3.6	Mississippi Valley Loess Plains	322	1.7%	29	0.4%			208	1.2%	2090	5.6%	327	1.2%
	8.3.7	South Central Plains	711	3.8%	24	0.4%			249	1.4%	6197	16.7%	13	0.0%
	8.3.8	East Central Texas Plains	191	1.0%	4	0.1%			173	1.0%	275	0.7%		
8.4	8.4.1	Ridge and Valley	262	1.4%	414	6.3%			883	5.1%	690	1.9%	1880	6.8%
1	8.4.2	Central Appalachians	52	0.3%	64	1.0%			47	0.3%	118	0.3%	3411	12.3%
	8.4.3	Western Allegheny Plateau	134	0.7%	454	6.9%			63	0.4%	15	0.0%	2714	9.8%

	NA 12		gree Me N=1 Assoc I N/S	<b>n ash</b> dian 0,S=6 N-U, S-↓ = 0.45	black Mediar Assoc N/S =	walnut n N=12 c N-U = 0.13	Utah ju Mediar Assoc N/S =	uniper n N=3 n N-U 0.72	east redc Mec N=11 Assoc N N/S Co	eern edar lian ,S=7 I-U,S-↓ r= 0.3	swee Median I Assoc N N/S	etgum N=9, S=7 N-U, S-↓ = 0.37	<b>yellow p</b> Median Assoc N/S =	o <b>plar</b> S=11 S-↓ 0.4
L2	CODE	US L3NAME	count	%	count	%	count	%	count	%	count	%	count	%
	8.4.4	Blue Ridge	61	0.3%	82	1.2%			40	0.2%	85	0.2%	3203	11.6%
İ	8.4.5	Ozark Highlands	176	0.9%	810	12.3%			3903	22.6%	168	0.5%	4	0.0%
1	8.4.6	Boston Mountains	21	0.1%	35	0.5%			333	1.9%	218	0.6%		
	8.4.7	Arkansas Valley	166	0.9%	10	0.2%			727	4.2%	268	0.7%		
İ	8.4.8	Ouachita Mountains	101	0.5%	5	0.1%			260	1.5%	386	1.0%		
1	8.4.9	Southwestern Appalachians	154	0.8%	49	0.7%			415	2.4%	752	2.0%	1154	4.2%
8.5	8.5.1	Middle Atlantic Coastal Plain	530	2.8%	14	0.2%			21	0.1%	3130	8.4%	551	2.0%
İ.	8.5.2	Mississippi Alluvial Plain	1109	5.9%	9	0.1%			22	0.1%	877	2.4%	19	0.1%
Ì	8.5.3	Southern Coastal Plain	630	3.3%					43	0.2%	1422	3.8%	55	0.2%
ľ	8.5.4	Atlantic Coastal Pine Barrens	1	0.0%	3	0.0%			16	0.1%	85	0.2%	19	0.1%
9.2	9.2.1	Northern Glaciated Plains	383	2.0%					1	0.0%				
	9.2.2	Lake Agassiz Plain	280	1.5%										
Ì	9.2.3	Western Corn Belt Plains	502	2.7%	381	5.8%			384	2.2%				
ľ	9.2.4	Central Irregular Plains	408	2.2%	701	10.6%			435	2.5%				
9.3	9.3.1	Northwestern Glaciated Plains	96	0.5%					130	0.8%				+
Ì	9.3.3	Northwestern Great Plains	417	2.2%	1	0.0%			95	0.6%				
Ì	9.3.4	Nebraska Sand Hills	29	0.2%					104	0.6%				
9.4	9.4.1	High Plains	27	0.1%	1	0.0%			33	0.2%				+
İ.	9.4.2	Central Great Plains	337	1.8%	27	0.4%			319	1.8%				
	9.4.3	Southwestern Tablelands	12	0.1%	7	0.1%			8	0.0%				
ĺ	9.4.4	Flint Hills	50	0.3%	50	0.8%			73	0.4%				1
	9.4.5	Cross Timbers	25	0.1%	13	0.2%			19	0.1%				

			gree Me N=1 Assoc I N/S	e <b>n ash</b> edian 0,S=6 N-U, S-↓ = 0.45	black v Median Assoc N/S =	<b>valnut</b> i N=12 : N-U : 0.13	Utah ju Mediai Assoc N/S =	uniper n N=3 c N-U : 0.72	east redc Med N=11 Assoc N N/S Col	<b>ern</b> edar lian ,S=7 I-U,S-↓ r= 0.3	swee Median I Assoc N N/S	<b>tgum</b> N=9, S=7 N-U, S-↓ = 0.37	yellow p Median Assoc N/S =	<b>oplar</b> S=11 S-↓ 0.4
L2	CODE	US_L3NAME	count	%	count	%	count	%	count	%	count	%	count	%
	9.4.6	Edwards Plateau												
	9.4.7	Texas Blackland Prairies	1	0.0%					13	0.1%				
9.5	9.5.1	Western Gulf Coastal Plain	39	0.2%					11	0.1%	118	0.3%		
9.6	9.6.1	Southern Texas Plains												
10.1	10.1.2	Columbia Plateau												
	10.1.3	Northern Basin and Range					605	3.2%						
	10.1.4	Wyoming Basin					189	1.0%						
1	10.1.5	Central Basin and Range					4944	26.5%						
	10.1.6	Colorado Plateaus					7696	41.2%						
	10.1.7	Arizona/New Mexico Plateau					2211	11.8%						
1	10.1.8	Snake River Plain												
10.2	10.2.1	Mojave Basin and Range					207	1.1%						
	10.2.2	Sonoran Basin and Range					2	0.0%						
	10.2.10	Chihuahuan Deserts												
11.1	11.1.1	Southern and Central California Chaparral and Oak Woodlands												
1	11.1.2	Central California Valley												
1	11.1.3	Southern California Mountains					-							
12.1	12.1.1	Madrean Archipelago					2	0.0%						
13.1	13.1.1	Arizona/New Mexico Mountains					1457	7.8%						
15.4	15.4.1	Southern Florida Coastal Plain	7	0.0%										
		Total Tree Count	18854		6591		18681		17249		37211		27701	

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NA			swee Media Asso N/S =	e <b>tbay</b> in S=7 c S-↓ = 0.35	swamp Mediar Assoc N/S =	<b>tupelo</b> n S=6 s S-↓ 0.46	black Median Assoc N/S =	<b>gum</b> N=10 N-U 0.42	eas hophoi Media Asso N/S =	<b>tern</b> rnbeam In S=6 c S-↓ = 0.36	sour Median N Assoc N N/S	wood  =9, S=10 I-U, S-↓ = 0.3	<b>shortlea</b> Median Assoc N/S =	<b>f pine</b> S=6 S-↓ 0.18
L2	CODE	US_L3NAME	count	%	count	%	count	%	count	%	count	%	count	%
5.2	5.2.1	Northern Lakes and Forests					2	0.0%	1051	17.8%				
	5.2.2	Northern Minnesota Wetlands							5	0.1%				
5.3	5.3.1	Northeastern Highlands					29	0.2%	703	11.9%				
	5.3.3	North Central Appalachians					171	1.3%	116	2.0%				
6.2	6.2.3	Northern Rockies												
	6.2.4	Canadian Rockies												
	6.2.5	North Cascades												
	6.2.7	Cascades												
	6.2.8	Eastern Cascades Slopes and Foothills												
	6.2.9	Blue Mountains												
	6.2.10	Middle Rockies							3	0.1%				
	6.2.11	Klamath Mountains												
	6.2.12	Sierra Nevada												
	6.2.13	Wasatch and Uinta Mountains												
	6.2.14	Southern Rockies												
	6.2.15	Idaho Batholith												
7.1	7.1.7	Puget Lowland												
	7.1.8	Coast Range												
	7.1.9	Willamette Valley												
8.1	8.1.1	Eastern Great Lakes Lowlands					32	0.2%	241	4.1%				
	8.1.3	Northern Allegheny Plateau					16	0.1%	432	7.3%				

			swe Medi Asso N/S	etbay an S=7 oc S-↓ = 0.35	swamp Media Assoc N/S =	<b>tupelo</b> n S=6 c S-↓ = 0.46	black Mediar Assoc N/S =	<b>xgum</b> n N=10 c N-U = 0.42	eas hophor Media Asso N/S :	<b>tern</b> r <b>nbeam</b> in S=6 c S-↓ = 0.36	sour Median N Assoc N N/S	<b>wood</b> I=9, S=10 I-U, S-↓ = 0.3	shortlea Mediar Assoc N/S =	<b>af pine</b> a S=6 sS-↓ 0.18
NA_ L2	NA_L3 CODE	US_L3NAME	count	%	count	%	count	%	count	%	count	%	count	%
	8.1.4	North Central Hardwood Forests							560	9.5%				
	8.1.5	Driftless Area							454	7.7%				
	8.1.6	Southern Michigan/Northern Indiana Drift Plains					43	0.3%	68	1.2%				
	8.1.7	Northeastern Coastal Zone					121	0.9%	34	0.6%				
	8.1.8	Acadian Plains and Hills							231	3.9%				
	8.1.10	Erie Drift Plain					72	0.5%	84	1.4%	1	0.0%		
8.2	8.2.1	Southeastern Wisconsin Till Plains							34	0.6%				
	8.2.2	Huron/Erie Lake Plains					37	0.3%	18	0.3%				
	8.2.3	Central Corn Belt Plains					40	0.3%	13	0.2%				
	8.2.4	Eastern Corn Belt Plains					47	0.3%	78	1.3%				
8.3	8.3.1	Northern Piedmont					179	1.3%	4	0.1%	6	0.1%	8	0.0%
	8.3.2	Interior River Valleys and Hills					214	1.6%	83	1.4%	26	0.3%	27	0.2%
	8.3.3	Interior Plateau			3	0.0%	885	6.6%	201	3.4%	673	7.5%	276	1.6%
	8.3.4	Piedmont	42	1.0%	79	0.7%	957	7.1%	85	1.4%	1953	21.8%	2761	16.2%
	8.3.5	Southeastern Plains	2345	55.8%	4949	44.5%	2415	17.9%	239	4.0%	678	7.6%	1941	11.4%
	8.3.6	Mississippi Valley Loess Plains	12	0.3%	10	0.1%	164	1.2%	176	3.0%	51	0.6%	141	0.8%
	8.3.7	South Central Plains	262	6.2%	78	0.7%	1232	9.2%	309	5.2%	6	0.1%	1891	11.1%
	8.3.8	East Central Texas Plains					42	0.3%					30	0.2%
8.4	8.4.1	Ridge and Valley	3	0.1%	1	0.0%	1668	12.4%	121	2.0%	804	9.0%	383	2.2%
	8.4.2	Central Appalachians					606	4.5%	44	0.7%	845	9.4%	47	0.3%
	8.4.3	Western Allegheny Plateau					586	4.4%	100	1.7%	504	5.6%	65	0.4%
	8.4.4	Blue Ridge					1029	7.6%	50	0.8%	2363	26.4%	376	2.2%

			swe Medi Asso N/S	<b>etbay</b> an S=7 oc S-↓ = 0.35	swamp Media Asso N/S =	n S=6 c S-↓ = 0.46	black Mediar Assoc N/S =	<b>xgum</b> n N=10 c N-U = 0.42	eas hophor Media Asso N/S :	<b>tern</b> rnbeam an S=6 c S-↓ = 0.36	sour Median N Assoc N N/S	<b>wood</b> I=9, S=10 I-U, S-↓ = 0.3	shortlea Mediar Assoc N/S =	afpine n S=6 : S-↓ 0.18
L2	CODE	US_L3NAME	count	%	count	%	count	%	count	%	count	%	count	%
	8.4.5	Ozark Highlands					660	4.9%	12	0.2%			2867	16.8%
	8.4.6	Boston Mountains					333	2.5%	10	0.2%			679	4.0%
	8.4.7	Arkansas Valley			5	0.0%	112	0.8%	10	0.2%			1259	7.4%
	8.4.8	Ouachita Mountains					288	2.1%	43	0.7%			3804	22.3%
	8.4.9	Southwestern Appalachians					547	4.1%	36	0.6%	989	11.1%	381	2.2%
8.5	8.5.1	Middle Atlantic Coastal Plain	225	5.4%	2181	19.6%	506	3.8%	11	0.2%	43	0.5%	31	0.2%
	8.5.2	Mississippi Alluvial Plain			65	0.6%	33	0.2%	13	0.2%	3	0.0%	16	0.1%
	8.5.3	Southern Coastal Plain	1293	30.8%	3738	33.6%	278	2.1%	17	0.3%	1	0.0%	2	0.0%
	8.5.4	Atlantic Coastal Pine Barrens	10	0.2%			99	0.7%					40	0.2%
9.2	9.2.1	Northern Glaciated Plains							10	0.2%				
	9.2.2	Lake Agassiz Plain							11	0.2%				
	9.2.3	Western Corn Belt Plains							160	2.7%				
	9.2.4	Central Irregular Plains							36	0.6%				
9.3	9.3.1	Northwestern Glaciated Plains							2	0.0%				
	9.3.3	Northwestern Great Plains							2	0.0%				
	9.3.4	Nebraska Sand Hills												
9.4	9.4.1	High Plains												
	9.4.2	Central Great Plains												
	9.4.3	Southwestern Tablelands												
	9.4.4	Flint Hills							1	0.0%				
	9.4.5	Cross Timbers											3	0.0%
	9.4.6	Edwards Plateau												

			swe Media Asso N/S	<b>etbay</b> an S=7 oc S-↓ = 0.35	swamp Median Assoc N/S =	<b>tupelo</b> n S=6 c S-↓ ≤ 0.46	black Median Assoc N/S =	<b>gum</b> N=10 N-U 0.42	eas hophor Media Asso N/S :	<b>tern</b> r <b>nbeam</b> in S=6 c S-↓ = 0.36	sour Median N Assoc N N/S	<b>wood</b> I=9, S=10 I-U, S-↓ = 0.3	<b>shortlea</b> Median Assoc N/S =	<b>f pine</b> S=6 S-↓ 0.18
L2	CODE	US_L3NAME	count	%	count	%	count	%	count	%	count	%	count	%
	9.4.7	Texas Blackland Prairies												
9.5	9.5.1	Western Gulf Coastal Plain	1	0.0%			21	0.2%	1	0.0%				
9.6	9.6.1	Southern Texas Plains												
10.1	10.1.2	Columbia Plateau												
	10.1.3	Northern Basin and Range												
	10.1.4	Wyoming Basin												
	10.1.5	Central Basin and Range												
	10.1.6	Colorado Plateaus												
	10.1.7	Arizona/New Mexico Plateau												
	10.1.8	Snake River Plain												
10.2	10.2.1	Mojave Basin and Range												
	10.2.2	Sonoran Basin and Range												
	10.2.10	Chihuahuan Deserts												
11.1	11.1.1	Southern and Central California Chaparral and Oak Woodlands												
	11.1.2	Central California Valley												
	11.1.3	Southern California Mountains												
12.1	12.1.1	Madrean Archipelago												
13.1	13.1.1	Arizona/New Mexico Mountains												
15.4	15.4.1	Southern Florida Coastal Plain	6	0.1%	1	0.0%								
		Total Tree Count	4199		11110		13464		5912		8946		17028	

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NA	NA 12		slasi Median Assoc I N/S	n <b>pine</b> N=7, S=5 N-U, S-↓ = 0.44	longlea Mediar Assoc N/S =	af pine n S=7 c S-↓ ≤ 0.44	red p Median N Assoc N N/S =	<b>bine</b> I=8, S=5 I-U, S-↓ ∺ 0.54	pitch Med N=10, Assoc N N/S =	<b>pine</b> ian S=12 I-U,S-↓ 0.65	eastern v Media Asso N/S	<b>vhite pine</b> an S=6 c S-↓ = 0.6	<b>Iobiolly</b> Median Assoc N/S = 0	<b>pine</b> S=7 S-↓ 0.32
L2	CODE	US L3NAME	count	%	Count	%	count	%	count	%	count	%	count	%
5.2	5.2.1	Northern Lakes and Forests					6593	65.0%			4340	18.4%		
1	5.2.2	Northern Minnesota Wetlands					208	2.1%			38	0.2%		
5.3	5.3.1	Northeastern Highlands					189	1.9%	88	2.8%	4501	19.1%		
	5.3.3	North Central Appalachians					44	0.4%	47	1.5%	566	2.4%		
6.2	6.2.3	Northern Rockies												
	6.2.4	Canadian Rockies												
1	6.2.5	North Cascades												
1	6.2.7	Cascades												
1	6.2.8	Eastern Cascades Slopes and Foothills												
1	6.2.9	Blue Mountains												
1	6.2.10	Middle Rockies												
1	6.2.11	Klamath Mountains												
1	6.2.12	Sierra Nevada												
1	6.2.13	Wasatch and Uinta Mountains												
1	6.2.14	Southern Rockies												
	6.2.15	Idaho Batholith							-					
7.1	7.1.7	Puget Lowland												
1	7.1.8	Coast Range												
1	7.1.9	Willamette Valley												
8.1	8.1.1	Eastern Great Lakes Lowlands					79	0.8%	7	0.2%	602	2.6%		
	8.1.3	Northern Allegheny Plateau					155	1.5%	3	0.1%	955	4.1%		

Ioblolly pine Median S=7 Assoc S-↓ N/S = 0.32	white pine an S=6 oc S-↓ = 0.6	eastern v Media Asso N/S	<b>pine</b> ian S=12 I-U,S-↓ 0.65	pitch Med N=10, Assoc N N/S =	<b>oine</b> I=8, S=5 -U, S-↓ 0.54	red p Median N Assoc N N/S =	<b>af pine</b> n S=7 c S-↓ ⊧ 0.44	longlea Mediar Assoc N/S =	n <b>pine</b> N=7, S=5 N-U, S-↓ = 0.44	slasł Median I Assoc I N/S			
count %	%	count	%	count	%	count	%	Count	%	count	US L3NAME	_ NA_L3 CODE	L2
	7.4%	1751			15.9%	1609					North Central Hardwood Forests	8.1.4	
	1.9%	436			3.4%	346					Driftless Area	8.1.5	
	1.5%	362	0.1%	2	4.6%	466					Southern Michigan/Northern Indiana Drift Plains	8.1.6	
	11.5%	2711	3.4%	106	0.1%	13					Northeastern Coastal Zone	8.1.7	
	10.0%	2353	0.2%	5	1.6%	163					Acadian Plains and Hills	8.1.8	
	0.1%	33			0.2%	20					Erie Drift Plain	8.1.10	
	0.6%	140			0.8%	85					Southeastern Wisconsin Till Plains	8.2.1	8.2
	0.5%	128			0.6%	60					Huron/Erie Lake Plains	8.2.2	
	0.1%	29			0.2%	19					Central Corn Belt Plains	8.2.3	
	0.2%	47			0.1%	13					Eastern Corn Belt Plains	8.2.4	
33 0.0%	0.3%	69									Northern Piedmont	8.3.1	8.3
40 0.1%	0.1%	16	0.2%	5	0.3%	29					Interior River Valleys and Hills	8.3.2	
578 0.8%	0.4%	106	0.2%	7	0.0%	2					Interior Plateau	8.3.3	
15095 21.8%	1.7%	401	0.8%	25			4.2%	224	0.2%	29	Piedmont	8.3.4	
23675 34.2%	0.0%	1	0.0%	1			57.8%	3108	33.9%	4035	Southeastern Plains	8.3.5	
1599 2.3%							0.0%	1	0.0%	3	Mississippi Valley Loess Plains	8.3.6	
13971 20.2%							5.8%	311	2.0%	235	South Central Plains	8.3.7	
151 0.2%									0.3%	37	East Central Texas Plains	8.3.8	
1494 2.2%	5.6%	1308	14.9%	472	0.1%	7	1.9%	100			Ridge and Valley	8.4.1	8.4
4 0.0%	1.0%	228	2.5%	80	0.1%	13					Central Appalachians	8.4.2	
100 0.1%	1.7%	391	3.4%	107	0.2%	24					Western Allegheny Plateau	8.4.3	
304 0.4%	7.7%	1817	9.9%	312	<u> </u>						Blue Ridge	8.4.4	
	0.6% 0.5% 0.1% 0.2% 0.3% 0.1% 0.4% 1.7% 0.0% 5.6% 1.0% 1.7% 7.7%	140 128 29 47 69 16 106 401 1 1 1308 228 391 1817	0.2% 0.2% 0.8% 0.0% 14.9% 2.5% 3.4% 9.9%	5 7 25 1 472 80 107 312	0.8% 0.6% 0.2% 0.1% 0.3% 0.0% 0.0% 0.1% 0.1% 0.2%	85 60 19 13 29 2 2 2 7 13 24	4.2% 57.8% 0.0% 5.8% 1.9%	224 3108 1 311 100	0.2% 33.9% 0.0% 2.0% 0.3%	29 4035 3 235 37	Southeastern Wisconsin Till Plains Huron/Erie Lake Plains Central Corn Belt Plains Eastern Corn Belt Plains Northern Piedmont Interior River Valleys and Hills Interior Plateau Piedmont Southeastern Plains Mississippi Valley Loess Plains South Central Plains East Central Texas Plains Ridge and Valley Central Appalachians Western Allegheny Plateau Blue Ridge	8.2.1         8.2.2         8.2.3         8.2.4         8.3.1         8.3.2         8.3.3         8.3.4         8.3.5         8.3.6         8.3.7         8.3.8         8.4.1         8.4.2         8.4.3         8.4.4	8.2 8.3 8.4

	NA 12		slas Median Assoc N/S	<b>h pine</b> N=7, S=5 N-U, S-↓ = 0.44	longlea Media Assoc N/S =	<b>af pine</b> n S=7 c S-↓ ∈ 0.44	red µ Median N Assoc N N/S =	<b>bine</b> I=8, S=5 -U, S-↓ ≆ 0.54	pitch Mea N=10 Assoc N N/S =	<b>pine</b> dian ,S=12 N-U,S-↓ = 0.65	eastern v Media Asso N/S	white pine an S=6 ic S-↓ = 0.6	loblolly Median Assoc N/S =	r <b>pine</b> S=7 S-↓ 0.32
L2	CODE	US L3NAME	count	%	Count	%	count	%	count	%	count	%	count	%
	8.4.5	Ozark Highlands											21	0.0%
	8.4.6	Boston Mountains											50	0.1%
	8.4.7	Arkansas Valley											205	0.3%
	8.4.8	Ouachita Mountains											715	1.0%
	8.4.9	Southwestern Appalachians			9	0.2%			27	0.9%	130	0.6%	1486	2.1%
8.5	8.5.1	Middle Atlantic Coastal Plain	120	1.0%	353	6.6%			42	1.3%			7035	10.1%
	8.5.2	Mississippi Alluvial Plain											103	0.1%
	8.5.3	Southern Coastal Plain	7160	60.2%	1258	23.4%							2279	3.3%
	8.5.4	Atlantic Coastal Pine Barrens							1827	57.8%	103	0.4%	2	0.0%
9.2	9.2.1	Northern Glaciated Plains												
	9.2.2	Lake Agassiz Plain												
	9.2.3	Western Corn Belt Plains												
	9.2.4	Central Irregular Plains					2	0.0%						
9.3	9.3.1	Northwestern Glaciated Plains												
	9.3.3	Northwestern Great Plains												
	9.3.4	Nebraska Sand Hills												
9.4	9.4.1	High Plains					İ							
	9.4.2	Central Great Plains												
	9.4.3	Southwestern Tablelands												
	9.4.4	Flint Hills	1											1
1	9.4.5	Cross Timbers	1											1
	9.4.6	Edwards Plateau												

			<b>slas</b> Median Assoc N/S	<b>h pine</b> N=7, S=5 N-U, S-↓ = 0.44	longlea Media Assoc N/S =	a <b>fpine</b> n S=7 c S-↓ ⊧ 0.44	red µ Median N Assoc N N/S =	<b>bine</b> I=8, S=5 I-U, S-↓ ∺ 0.54	pitch Med N=10, Assoc N N/S =	<b>pine</b> lian S=12 I-U,S-↓ 0.65	eastern v Media Asso N/S	white pine an S=6 ic S-↓ = 0.6	loblolly Median Assoc N/S =	<b>pine</b> S=7 S-↓ 0.32
L2	CODE	US_L3NAME	count	%	Count	%	count	%	count	%	count	%	count	%
	9.4.7	Texas Blackland Prairies											75	0.1%
9.5	9.5.1	Western Gulf Coastal Plain			9	0.2%							306	0.4%
9.6	9.6.1	Southern Texas Plains												
10.1	10.1.2	Columbia Plateau												
	10.1.3	Northern Basin and Range												
	10.1.4	Wyoming Basin												
	10.1.5	Central Basin and Range												
	10.1.6	Colorado Plateaus												
	10.1.7	Arizona/New Mexico Plateau									-			
	10.1.8	Snake River Plain												
10.2	10.2.1	Mojave Basin and Range												
	10.2.2	Sonoran Basin and Range												
	10.2.10	Chihuahuan Deserts									-			
11.1	11.1.1	Southern and Central California Chaparral and Oak Woodlands												
	11.1.2	Central California Valley												
	11.1.3	Southern California Mountains												
12.1	12.1.1	Madrean Archipelago												
13.1	13.1.1	Arizona/New Mexico Mountains												
15.4	15.4.1	Southern Florida Coastal Plain	283	2.4%										
		Total Tree Count	11902		5373		10139		3163		23562		69321	

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	NA 12		Virgir Me N=1( Assoc N/S	<b>tia pine</b> edian ),S=11 N-U, S-↓ = 0.44	bigtootl Median N Assoc N N/S =	h <b>aspen</b> \=9, S=6 I-U, S-↓ = 0.57	<b>quaking</b> Median N Assoc N N/S =	<b>g aspen</b> √=7, S=3 I-U, S-↓ = 0.61	black of Mediar Assoc N/S =	<b>cherry</b> n N=11 c N-U = 0.33	Dougl Media Assoc N/S =	a <b>s-fir</b> n N=3 c N-U : 0.65	white Median N Assoc N N/S =	<b>oak</b> =10, S=8 -U, S-↓ 0.17
L2	CODE	US_L3NAME	count	%	count	%	count	%	count	%	count	%	count	%
5.2	5.2.1	Northern Lakes and Forests			6784	58.8%	27494	52.9%	1989	8.1%			1615	3.4%
	5.2.2	Northern Minnesota Wetlands			13	0.1%	2995	5.8%						
5.3	5.3.1	Northeastern Highlands			496	4.3%	1273	2.5%	1539	6.3%			243	0.5%
	5.3.3	North Central Appalachians			115	1.0%	167	0.3%	1516	6.2%			1032	2.2%
6.2	6.2.3	Northern Rockies					67	0.1%			4463	9.4%		
	6.2.4	Canadian Rockies					92	0.2%			800	1.7%		
	6.2.5	North Cascades									2702	5.7%		
	6.2.7	Cascades									10680	22.5%		
	6.2.8	Eastern Cascades Slopes and Foothills					28	0.1%			1631	3.4%		
	6.2.9	Blue Mountains					7	0.0%			3271	6.9%		
	6.2.10	Middle Rockies					398	0.8%			3908	8.2%		
	6.2.11	Klamath Mountains					3	0.0%			6885	14.5%		
	6.2.12	Sierra Nevada					30	0.1%			1066	2.2%		
	6.2.13	Wasatch and Uinta Mountains					3024	5.8%			774	1.6%		
	6.2.14	Southern Rockies					5182	10.0%			2161	4.6%		
	6.2.15	Idaho Batholith					15	0.0%			1699	3.6%		
7.1	7.1.7	Puget Lowland									409	0.9%		
	7.1.8	Coast Range									4823	10.2%		
	7.1.9	Willamette Valley									184	0.4%		
8.1	8.1.1	Eastern Great Lakes Lowlands			88	0.8%	384	0.7%	408	1.7%			27	0.1%
	8.1.3	Northern Allegheny Plateau			166	1.4%	428	0.8%	867	3.5%	9	0.0%	180	0.4%

			Virgir Me N=10 Assoc N/S	<b>nia pine</b> edian ),S=11 N-U, S-↓ = 0.44	bigtooth Median N Assoc N N/S =	n <b>aspen</b> √=9, S=6 I-U, S-↓ ⊧ 0.57	<b>quaking</b> Median N Assoc N N/S =	<b>J aspen</b> I=7, S=3 I-U, S-↓ ∺ 0.61	black of Median Assoc N/S =	<b>cherry</b> N=11 N-U 0.33	Dougl Mediau Assoc N/S =	<b>as-fir</b> n N=3 c N-U c 0.65	white Median N= Assoc N- N/S =	<b>oak</b> =10, S=8 -U, S-↓ 0.17
L2	NA_L3	US_L3NAME	count	%	count	%	count	%	count	%	count	%	count	%
	8.1.4	North Central Hardwood Forests			1026	8.9%	4024	7.7%	692	2.8%			1060	2.3%
	8.1.5	Driftless Area			540	4.7%	488	0.9%	894	3.7%			1247	2.7%
	8.1.6	Southern Michigan/Northern Indiana Drift Plains			396	3.4%	344	0.7%	1508	6.2%	1	0.0%	749	1.6%
	8.1.7	Northeastern Coastal Zone			157	1.4%	184	0.4%	312	1.3%			817	1.7%
	8.1.8	Acadian Plains and Hills			612	5.3%	1183	2.3%	169	0.7%			17	0.0%
	8.1.10	Erie Drift Plain			105	0.9%	202	0.4%	879	3.6%			74	0.2%
8.2	8.2.1	Southeastern Wisconsin Till Plains			30	0.3%	296	0.6%	490	2.0%			164	0.3%
	8.2.2	Huron/Erie Lake Plains			200	1.7%	421	0.8%	154	0.6%			97	0.2%
	8.2.3	Central Corn Belt Plains	1	0.0%			21	0.0%	350	1.4%			128	0.3%
	8.2.4	Eastern Corn Belt Plains	3	0.0%	21	0.2%	8	0.0%	603	2.5%			177	0.4%
8.3	8.3.1	Northern Piedmont	238	2.6%	18	0.2%	6	0.0%	283	1.2%	9	0.0%	328	0.7%
	8.3.2	Interior River Valleys and Hills	78	0.8%	9	0.1%			578	2.4%			1594	3.4%
	8.3.3	Interior Plateau	339	3.6%	30	0.3%			976	4.0%			3240	6.9%
	8.3.4	Piedmont	3590	38.5%	9	0.1%			970	4.0%			4634	9.9%
	8.3.5	Southeastern Plains	412	4.4%	7	0.1%			1163	4.7%			2853	6.1%
	8.3.6	Mississippi Valley Loess Plains							258	1.1%			448	1.0%
	8.3.7	South Central Plains	1	0.0%					157	0.6%			1221	2.6%
	8.3.8	East Central Texas Plains							4	0.0%				
8.4	8.4.1	Ridge and Valley	1653	17.7%	111	1.0%	24	0.0%	1398	5.7%	1	0.0%	2923	6.2%
	8.4.2	Central Appalachians	212	2.3%	101	0.9%	27	0.1%	1396	5.7%			1976	4.2%
	8.4.3	Western Allegheny Plateau	574	6.2%	505	4.4%	109	0.2%	2712	11.1 %			2442	5.2%

				Virginia pine Median N=10,S=11 Assoc N-U, S-↓ N/S = 0.44		bigtooth aspen Median N=9, S=6 Assoc N-U, S-↓ N/S = 0.57		<b>quaking aspen</b> Median N=7, S=3 Assoc N-U, S-↓ N/S = 0.61		black cherry Median N=11 Assoc N-U N/S = 0.33		Douglas-fir Median N=3 Assoc N-U N/S = 0.65		white oak Median N=10, S=8 Assoc N-U, S-↓ N/S = 0.17	
	NA_L3 CODE	US_L3NAME	count	%	count	%	count	%	count	%	count	%	count	%	
	8.4.4	Blue Ridge	946	10.1%	1	0.0%			417	1.7%			1265	2.7%	
	8.4.5	Ozark Highlands	3	0.0%					448	1.8%			8989	19.2%	
	8.4.6	Boston Mountains							96	0.4%			1619	3.5%	
	8.4.7	Arkansas Valley							73	0.3%			487	1.0%	
	8.4.8	Ouachita Mountains							106	0.4%			1268	2.7%	
	8.4.9	Southwestern Appalachians	1155	12.4%					260	1.1%			2152	4.6%	
8.5	8.5.1	Middle Atlantic Coastal Plain	83	0.9%	2	0.0%			250	1.0%			458	1.0%	
	8.5.2	Mississippi Alluvial Plain							40	0.2%			83	0.2%	
	8.5.3	Southern Coastal Plain	14	0.2%					85	0.3%			13	0.0%	
	8.5.4	Atlantic Coastal Pine Barrens	22	0.2%	3	0.0%			40	0.2%			519	1.1%	
9.2	9.2.1	Northern Glaciated Plains					377	0.7%							
	9.2.2	Lake Agassiz Plain			1	0.0%	1773	3.4%	3	0.0%					
	9.2.3	Western Corn Belt Plains			1	0.0%	86	0.2%	210	0.9%			112	0.2%	
	9.2.4	Central Irregular Plains							192	0.8%			676	1.4%	
9.3	9.3.1	Northwestern Glaciated Plains					10	0.0%			12	0.0%			
	9.3.3	Northwestern Great Plains					30	0.1%	1	0.0%	260	0.5%			
	9.3.4	Nebraska Sand Hills													
9.4	9.4.1	High Plains													
	9.4.2	Central Great Plains													
	9.4.3	Southwestern Tablelands													
	9.4.4	Flint Hills													
	9.4.5	Cross Timbers							2	0.0%					

NA		2		Virginia pine Median         big Meci Meci Ass Ass N/S = 0.44		<b>igtooth aspen</b> edian N=9, S=6 \ssoc N-U, S-↓ N/S = 0.57		quaking aspen Median N=7, S=3 Assoc N-U, S-↓ N/S = 0.61		black cherry Median N=11 Assoc N-U N/S = 0.33		Douglas-fir Median N=3 Assoc N-U N/S = 0.65		white oak Median N=10, S=8 Assoc N-U, S-↓ N/S = 0.17	
L2	CODE	US_L3NAME	count	%	count	%	count	%	count	%	count	%	count	%	
	9.4.6	Edwards Plateau													
	9.4.7	Texas Blackland Prairies													
9.5	9.5.1	Western Gulf Coastal Plain							5	0.0%					
9.6	9.6.1	Southern Texas Plains													
10.1	10.1.2	Columbia Plateau					37	0.1%			174	0.4%			
	10.1.3	Northern Basin and Range					112	0.2%			105	0.2%			
	10.1.4	Wyoming Basin									27	0.1%			
	10.1.5	Central Basin and Range					62	0.1%			26	0.1%			
	10.1.6	Colorado Plateaus					282	0.5%			403	0.8%			
	10.1.7	Arizona/New Mexico Plateau					3	0.0%			17	0.0%			
	10.1.8	Snake River Plain					32	0.1%							
10.2	10.2.1	Mojave Basin and Range													
	10.2.2	Sonoran Basin and Range													
	10.2.10	Chihuahuan Deserts													
11.1	11.1.1	Southern and Central California Chaparral and Oak Woodlands									222	0.5%			
	11.1.2	Central California Valley													
	11.1.3	Southern California Mountains													
12.1	12.1.1	Madrean Archipelago									38	0.1%			
13.1	13.1.1	Arizona/New Mexico Mountains					218	0.4%			657	1.4%			
15.4	15.4.1	Southern Florida Coastal Plain													
		Total Tree Count	9324		11547		51946		24493		47417		46927		

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ΝΑ	NA 12		scarlet oak Median N=10,S=10 Assoc N-↓, S-↓ N/S = 0.37		southern red oak Median N=9 Assoc N-↓ N/S = 0.36		<b>laurel oak</b> Median S=6 Assoc S-↓ N/S = 0.41		<b>chinkapin oak</b> Median N=11 Assoc N-U N/S = 0.31		water oak Median N=8, S=9 Assoc N-↓, S-↓ N/S = 0.26		chestnut oak Median S=12 Assoc S-↓ N/S = 0.44	
L2	CODE	US_L3NAME	count	%	count	%	count	%	count	%	count	%	count	%
5.2	5.2.1	Northern Lakes and Forests												
	5.2.2	Northern Minnesota Wetlands												
5.3	5.3.1	Northeastern Highlands	112	1.1%									386	1.6%
	5.3.3	North Central Appalachians	236	2.2%									1155	4.9%
6.2	6.2.3	Northern Rockies												
	6.2.4	Canadian Rockies												
	6.2.5	North Cascades												
	6.2.7	Cascades												
	6.2.8	Eastern Cascades Slopes and Foothills												
	6.2.9	Blue Mountains												
	6.2.10	Middle Rockies												
	6.2.11	Klamath Mountains												
	6.2.12	Sierra Nevada												
	6.2.13	Wasatch and Uinta Mountains												
	6.2.14	Southern Rockies												
	6.2.15	Idaho Batholith												
7.1	7.1.7	Puget Lowland												
	7.1.8	Coast Range							2	0.1%				
	7.1.9	Willamette Valley												
8.1	8.1.1	Eastern Great Lakes Lowlands											3	0.0%
	8.1.3	Northern Allegheny Plateau	21	0.2%									229	1.0%
			scar Me N=10 Assoc N/S	<b>let oak</b> edian ),S=10 N-↓, S-↓ = 0.37	southe oa Media Assoo N/S =	ern red ak n N=9 c N-↓ = 0.36	laure Media Assoc N/S =	<b>I oak</b> n S=6 c S-↓ ⊧ 0.41	<b>chinka</b> Mediar Assoc N/S =	<b>oin oak</b> N=11 N-U 0.31	wate Median I Assoc I N/S	<b>r oak</b> N=8, S=9 N-↓, S-↓ = 0.26	<b>chestn</b> Mediar Assoc N/S =	<b>ut oak</b> ì S=12 c S-↓ = 0.44
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L2	NA_L3	US_L3NAME	count	%	count	%	count	%	count	%	count	%	count	%
	8.1.4	North Central Hardwood Forests												
	8.1.5	Driftless Area							17	0.6%				
	8.1.6	Southern Michigan/Northern Indiana Drift Plains	7	0.1%					3	0.1%				
	8.1.7	Northeastern Coastal Zone	636	6.0%					4	0.1%			187	0.8%
	8.1.8	Acadian Plains and Hills												
	8.1.10	Erie Drift Plain	5	0.0%									5	0.0%
8.2	8.2.1	Southeastern Wisconsin Till Plains												
	8.2.2	Huron/Erie Lake Plains	5	0.0%					4	0.1%				
	8.2.3	Central Corn Belt Plains							9	0.3%				
	8.2.4	Eastern Corn Belt Plains	5	0.0%					104	3.4%			6	0.0%
8.3	8.3.1	Northern Piedmont	79	0.7%	90	1.0%							400	1.7%
	8.3.2	Interior River Valleys and Hills	72	0.7%	57	0.6%			275	8.9%			86	0.4%
	8.3.3	Interior Plateau	612	5.8%	498	5.6%	1	0.0%	1002	32.4 %	70	0.5%	1183	5.1%
	8.3.4	Piedmont	961	9.0%	1646	18.6%	52	0.9%	2	0.1%	1546	10.6%	1313	5.6%
	8.3.5	Southeastern Plains	326	3.1%	2263	25.6%	2724	46.9%	28	0.9%	6454	44.3%	269	1.1%
	8.3.6	Mississippi Valley Loess Plains	14	0.1%	278	3.1%	29	0.5%	22	0.7%	605	4.2%		
	8.3.7	South Central Plains	1	0.0%	1597	18.0%	341	5.9%	11	0.4%	2446	16.8%		
	8.3.8	East Central Texas Plains			181	2.0%	3	0.1%			203	1.4%		
8.4	8.4.1	Ridge and Valley	1707	16.0%	385	4.3%	17	0.3%	124	4.0%	149	1.0%	7903	33.7%
	8.4.2	Central Appalachians	874	8.2%	34	0.4%			44	1.4%			2496	10.7%
	8.4.3	Western Allegheny Plateau	586	5.5%	11	0.1%			53	1.7%			1305	5.6%

	NA 12		scar Me N=10 Assoc N/S	<b>et oak</b> dian ),S=10 N-↓, S-↓ = 0.37	southe oa Media Assoc N/S =	e <b>rn red</b> ak n N=9 c N-↓ = 0.36	laure Media Assoc N/S =	l <b>oak</b> n S=6 c S-↓ ≑ 0.41	<b>chinka</b> Mediar Assoc N/S =	<b>oin oak</b> n N=11 c N-U : 0.31	wate Median Assoc I N/S	e <b>r oak</b> N=8, S=9 N-↓, S-↓ = 0.26	<b>chestn</b> Mediar Assoc N/S =	<b>ut oak</b> n S=12 c S-↓ = 0.44
L2	CODE	US_L3NAME	count	%	count	%	count	%	count	%	count	%	count	%
	8.4.4	Blue Ridge	1749	16.4%	254	2.9%			5	0.2%	22	0.2%	4800	20.5%
	8.4.5	Ozark Highlands	1508	14.2%	538	6.1%	2	0.0%	848	27.5 %	1	0.0%		
	8.4.6	Boston Mountains			81	0.9%			75	2.4%				
	8.4.7	Arkansas Valley			156	1.8%	2	0.0%	3	0.1%	140	1.0%		
	8.4.8	Ouachita Mountains			130	1.5%			5	0.2%	76	0.5%		
	8.4.9	Southwestern Appalachians	586	5.5%	305	3.4%	2	0.0%	253	8.2%	82	0.6%	1590	6.8%
8.5	8.5.1	Middle Atlantic Coastal Plain	102	1.0%	211	2.4%	544	9.4%			1111	7.6%	25	0.1%
	8.5.2	Mississippi Alluvial Plain	1	0.0%	75	0.8%	20	0.3%	8	0.3%	286	2.0%		
	8.5.3	Southern Coastal Plain	1	0.0%	20	0.2%	2003	34.5%	2	0.1%	1233	8.5%		
	8.5.4	Atlantic Coastal Pine Barrens	432	4.1%	22	0.2%							84	0.4%
9.2	9.2.1	Northern Glaciated Plains												
	9.2.2	Lake Agassiz Plain												
	9.2.3	Western Corn Belt Plains							47	1.5%				
	9.2.4	Central Irregular Plains	2	0.0%	1	0.0%			96	3.1%				
9.3	9.3.1	Northwestern Glaciated Plains												
	9.3.3	Northwestern Great Plains												
	9.3.4	Nebraska Sand Hills												
9.4	9.4.1	High Plains												
	9.4.2	Central Great Plains							1	0.0%				
	9.4.3	Southwestern Tablelands												
	9.4.4	Flint Hills							34	1.1%				
	9.4.5	Cross Timbers							7	0.2%				

NA			scarl Me N=10 Assoc I N/S	<b>et oak</b> dian I,S=10 N-↓, S-↓ = 0.37	southe oa Mediar Assoc N/S =	<b>rn red</b> k n N=9 c N-↓ 0.36	laure Mediar Assoc N/S =	l <b>oak</b> n S=6 c S-↓ ≑ 0.41	chinkar Median Assoc N/S =	<b>oin oak</b> N=11 N-U 0.31	wate Median N Assoc N N/S =	<b>r oak</b> \=8, S=9 \-↓, S-↓ = 0.26	chestn Median Assoc N/S =	<b>ut oak</b> ⊧ S=12 ≳ S-↓ ≋ 0.44
L2	CODE	US_L3NAME	count	%	count	%	count	%	count	%	count	%	count	%
	9.4.6	Edwards Plateau												
	9.4.7	Texas Blackland Prairies			1	0.0%					3	0.0%		
9.5	9.5.1	Western Gulf Coastal Plain			21	0.2%	29	0.5%			139	1.0%		
9.6	9.6.1	Southern Texas Plains												
10.1	10.1.2	Columbia Plateau												
	10.1.3	Northern Basin and Range												
	10.1.4	Wyoming Basin												
	10.1.5	Central Basin and Range												
	10.1.6	Colorado Plateaus												
	10.1.7	Arizona/New Mexico Plateau												
	10.1.8	Snake River Plain												
10.2	10.2.1	Mojave Basin and Range												
	10.2.2	Sonoran Basin and Range												
	10.2.10	Chihuahuan Deserts												
11.1	11.1.1	Southern and Central California Chaparral and Oak Woodlands												
	11.1.2	Central California Valley												
	11.1.3	Southern California Mountains												
12.1	12.1.1	Madrean Archipelago												
13.1	13.1.1	Arizona/New Mexico Mountains							1	0.0%				
15.4	15.4.1	Southern Florida Coastal Plain					44	0.8%						
		Total Tree Count	10640		8855		5813		3089		14566		23425	

			north o Media Asso N/S	<b>ern red</b> p <b>ak</b> n N=10 pc N-U = 0.41	post Media Assoc N/S =	<b>oak</b> an 10 c N-U = 0.14	black Media Assoc N/S =	a <b>oak</b> n S=8 c S-↓ ⊧ 0.15	black Mec N=11, Assoc N N/S =	l <b>ocust</b> lian S=12 I-↑,S-↓ 0.19	sass Media Asso N/S	a <b>fras</b> n S=12 bc S-↓ = 0.3	baldcyp Median Assoc N/S =	n <b>ress</b> S=6 S-↓ 0.55
L2	CODE	US L3NAME	count	%	count	%	count	%	count	%	count	%	count	%
5.2	5.2.1	Northern Lakes and Forests	6123	19.3%			743	3.4%	28	0.5%	25	0.4%		
1	5.2.2	Northern Minnesota Wetlands	2	0.0%										<u> </u>
5.3	5.3.1	Northeastern Highlands	3162	10.0%			173	0.8%	34	0.6%	31	0.5%	1	
	5.3.3	North Central Appalachians	1104	3.5%			174	0.8%	12	0.2%	244	3.9%		
6.2	6.2.3	Northern Rockies			İ								1	<u> </u>
	6.2.4	Canadian Rockies					-		-				-	
1	6.2.5	North Cascades												<u> </u>
	6.2.7	Cascades												
1	6.2.8	Eastern Cascades Slopes and Foothills												<u> </u>
1	6.2.9	Blue Mountains					_							
1	6.2.10	Middle Rockies												
1	6.2.11	Klamath Mountains												
1	6.2.12	Sierra Nevada			<u> </u>									
1	6.2.13	Wasatch and Uinta Mountains												
	6.2.14	Southern Rockies			<u> </u>						-			
1	6.2.15	Idaho Batholith					-							
7.1	7.1.7	Puget Lowland												
1	7.1.8	Coast Range												
1	7.1.9	Willamette Valley												
8.1	8.1.1	Eastern Great Lakes Lowlands	258	0.8%			24	0.1%	30	0.5%	2	0.0%		
1	8.1.3	Northern Allegheny Plateau	886	2.8%	İ		123	0.6%	65	1.2%	9	0.1%	1	1

NA	NA 12		north o Media Asso N/S	ern red ak n N=10 bc N-U = 0.41	post Media Assoc N/S =	<b>oak</b> an 10 c N-U : 0.14	black Media Assoc N/S =	x <b>oak</b> n S=8 c S-↓ ÷ 0.15	black Mec N=11, Assoc N N/S =	lian S=12 V-↑,S-↓ 0.19	sass Media Assc N/S	a <b>fras</b> n S=12 bc S-↓ = 0.3	baldcyp Median Assoc N/S =	oress S=6 S-↓ 0.55
	CODE	US_L3NAME	count	%	count	%	count	%	count	%	count	%	count	%
	8.1.4	North Central Hardwood Forests	1604	5.1%			988	4.5%	143	2.6%				
1	8.1.5	Driftless Area	1673	5.3%			748	3.4%	128	2.3%				
1	8.1.6	Southern Michigan/Northern Indiana Drift Plains	724	2.3%			820	3.7%	137	2.5%	491	7.8%		
1	8.1.7	Northeastern Coastal Zone	1761	5.6%			911	4.2%	80	1.4%	46	0.7%		
1	8.1.8	Acadian Plains and Hills	868	2.7%			9	0.0%						
1	8.1.10	Erie Drift Plain	233	0.7%			60	0.3%	88	1.6%	60	1.0%		
8.2	8.2.1	Southeastern Wisconsin Till Plains	157	0.5%			64	0.3%	108	2.0%				
1	8.2.2	Huron/Erie Lake Plains	194	0.6%			25	0.1%	7	0.1%	63	1.0%		
1	8.2.3	Central Corn Belt Plains	73	0.2%			104	0.5%	93	1.7%	43	0.7%		
	8.2.4	Eastern Corn Belt Plains	250	0.8%	3	0.0%	107	0.5%	184	3.3%	148	2.4%		
8.3	8.3.1	Northern Piedmont	192	0.6%	10	0.0%	179	0.8%	103	1.9%	91	1.4%		
1	8.3.2	Interior River Valleys and Hills	536	1.7%	551	2.7%	819	3.7%	189	3.4%	600	9.6%	7	0.2%
1	8.3.3	Interior Plateau	793	2.5%	687	3.4%	1053	4.8%	520	9.4%	1078	17.2%	6	0.1%
1	8.3.4	Piedmont	901	2.8%	991	4.9%	773	3.5%	120	2.2%	68	1.1%	4	0.1%
1	8.3.5	Southeastern Plains	200	0.6%	1416	7.0%	445	2.0%	45	0.8%	141	2.2%	712	17.1%
1	8.3.6	Mississippi Valley Loess Plains	44	0.1%	172	0.8%	81	0.4%	51	0.9%	119	1.9%	88	2.1%
1	8.3.7	South Central Plains	7	0.0%	1673	8.3%	86	0.4%	14	0.3%	112	1.8%	643	15.4%
1	8.3.8	East Central Texas Plains			934	4.6%	4	0.0%	3	0.1%	19	0.3%		
8.4	8.4.1	Ridge and Valley	2603	8.2%	299	1.5%	1593	7.3%	826	14.9%	642	10.2%		
1	8.4.2	Central Appalachians	1336	4.2%	26	0.1%	678	3.1%	603	10.9%	524	8.3%		
1	8.4.3	Western Allegheny Plateau	952	3.0%	46	0.2%	1009	4.6%	605	10.9%	836	13.3%	1	0.0%
	8.4.4	Blue Ridge	1439	4.5%	88	0.4%	593	2.7%	752	13.6%	218	3.5%		

NA	NA 12		north Media Asso N/S	ern red pak in N=10 pc N-U = 0.41	post Media Assoc N/S =	<b>oak</b> an 10 c N-U = 0.14	black Media Assoc N/S =	x <b>oak</b> n S=8 c S-↓ ÷ 0.15	black Mec N=11, Assoc N N/S =	lian S=12 V-↑,S-↓ 0.19	sass Media Asso N/S	afras n S=12 bc S-↓ = 0.3	baldcyj Median Assoc N/S =	oress S=6 S-↓ 0.55
	CODE	US_L3NAME	count	%	count	%	count	%	count	%	count	%	count	%
	8.4.5	Ozark Highlands	1437	4.5%	6909	34.1%	7233	33.0%	31	0.6%	334	5.3%		
	8.4.6	Boston Mountains	741	2.3%	996	4.9%	527	2.4%	78	1.4%	36	0.6%		
	8.4.7	Arkansas Valley	188	0.6%	2263	11.2%	151	0.7%	6	0.1%	4	0.1%		
	8.4.8	Ouachita Mountains	451	1.4%	1902	9.4%	225	1.0%	5	0.1%	2	0.0%		
	8.4.9	Southwestern Appalachians	327	1.0%	276	1.4%	547	2.5%	70	1.3%	140	2.2%		
8.5	8.5.1	Middle Atlantic Coastal Plain	26	0.1%	63	0.3%	73	0.3%	29	0.5%	61	1.0%	456	11.0%
	8.5.2	Mississippi Alluvial Plain	8	0.0%	153	0.8%	13	0.1%	18	0.3%	17	0.3%	985	23.7%
	8.5.3	Southern Coastal Plain			39	0.2%	2	0.0%			1	0.0%	1044	25.1%
	8.5.4	Atlantic Coastal Pine Barrens	16	0.1%	31	0.2%	280	1.3%	10	0.2%	67	1.1%		
9.2	9.2.1	Northern Glaciated Plains			0	0.0%								
	9.2.2	Lake Agassiz Plain	2	0.0%	0	0.0%								
	9.2.3	Western Corn Belt Plains	192	0.6%	7	0.0%	84	0.4%	69	1.2%				
	9.2.4	Central Irregular Plains	220	0.7%	337	1.7%	336	1.5%	196	3.5%	4	0.1%		
9.3	9.3.1	Northwestern Glaciated Plains												
	9.3.3	Northwestern Great Plains												
	9.3.4	Nebraska Sand Hills												
9.4	9.4.1	High Plains												
	9.4.2	Central Great Plains							49	0.9%				
	9.4.3	Southwestern Tablelands							3	0.1%				
	9.4.4	Flint Hills	4	0.0%	3	0.0%								
	9.4.5	Cross Timbers	2	0.0%	372	1.8%	56	0.3%			2	0.0%		
1	9.4.6	Edwards Plateau			0	0.0%								

			north o Media Asso N/S	ern red pak n N=10 pc N-U = 0.41	post Media Assoc N/S =	<b>oak</b> an 10 c N-U = 0.14	black Media Assoc N/S =	x <b>oak</b> n S=8 c S-↓ ÷ 0.15	black   Med N=11, Assoc N N/S =	<b>ocust</b> lian S=12 I-↑,S-↓ 0.19	sass Media Asso N/S	afras n S=12 bc S-↓ = 0.3	baldcyp Median Assoc N/S =	<b>)ress</b> S=6 S-↓ 0.55
L2	CODE	US_L3NAME	count	%	count	%	count	%	count	%	count	%	count	%
	9.4.7	Texas Blackland Prairies			18	0.1%								
9.5	9.5.1	Western Gulf Coastal Plain			12	0.1%	1	0.0%	1	0.0%			29	0.7%
9.6	9.6.1	Southern Texas Plains												
10.1	10.1.2	Columbia Plateau												
	10.1.3	Northern Basin and Range												
	10.1.4	Wyoming Basin			<u> </u>								-	
	10.1.5	Central Basin and Range												
1	10.1.6	Colorado Plateaus												
	10.1.7	Arizona/New Mexico Plateau			<u> </u>								-	
1	10.1.8	Snake River Plain												
10.2	10.2.1	Mojave Basin and Range												
	10.2.2	Sonoran Basin and Range												
	10.2.10	Chihuahuan Deserts			<u> </u>								-	
11.1	11.1.1	Southern and Central California Chaparral and Oak Woodlands												
	11.1.2	Central California Valley												
	11.1.3	Southern California Mountains												
12.1	12.1.1	Madrean Archipelago												
13.1	13.1.1	Arizona/New Mexico Mountains												
15.4	15.4.1	Southern Florida Coastal Plain											187	4.5%
		Total Tree Count	31689		20277		21914		5533		6278		4162	

			Ame bass Media Asso N/S	erican swood an S=5 oc S-↓ = 0.39	east hem Media Assoc N/S =	<b>tern</b> Iock n N=8 c N-U = 0.78	winge Median Assoc N/S =	<b>d elm</b> N=10 N-↓ 0.37	Americ Mec N=11 Assoc N N/S =	<b>an elm</b> lian ,S=6 I-↓,S-↓ 0.24	slippe Median N Assoc I N/S	e <b>ry elm</b> J=11, S=8 N-U, S-↓ = 0.07	
L2	CODE	US L3NAME	count	%	count	%	count	%	count	%	count	%	
5.2	5.2.1	Northern Lakes and Forests	6603	43.4%	3441	13.4%			1340	7.0%	29	0.5%	
	5.2.2	Northern Minnesota Wetlands	155	1.0%					124	0.6%			
5.3	5.3.1	Northeastern Highlands	238	1.6%	7956	31.0%			233	1.2%	3	0.1%	
	5.3.3	North Central Appalachians	192	1.3%	1416	5.5%			23	0.1%	4	0.1%	
6.2	6.2.3	Northern Rockies											
1	6.2.4	Canadian Rockies											
	6.2.5	North Cascades											
	6.2.7	Cascades											
	6.2.8	Eastern Cascades Slopes and Foothills											
	6.2.9	Blue Mountains									-		
1	6.2.10	Middle Rockies							2	0.0%			
	6.2.11	Klamath Mountains											
	6.2.12	Sierra Nevada									-		
1	6.2.13	Wasatch and Uinta Mountains											
	6.2.14	Southern Rockies											
1	6.2.15	Idaho Batholith					-						
7.1	7.1.7	Puget Lowland											
1	7.1.8	Coast Range											
	7.1.9	Willamette Valley											
8.1	8.1.1	Eastern Great Lakes Lowlands	343	2.3%	1010	3.9%			508	2.7%	16	0.3%	1
	8.1.3	Northern Allegheny Plateau	398	2.6%	2257	8.8%			136	0.7%	1	0.0%	

NA	NA 12		Ame bass Media Asso N/S	erican swood an S=5 oc S-↓ = 0.39	east hem Media Assoc N/S =	ern lock n N=8 c N-U 0.78	winge Median Assoc N/S =	<b>d elm</b> n N=10 c N-↓ = 0.37	Americ Mec N=11 Assoc N N/S =	<b>an elm</b> lian ,S=6 I-↓,S-↓ 0.24	slippe Median N Assoc N N/S	e <b>ry elm</b> I=11, S=8 N-U, S-↓ = 0.07	
L2	CODE	US L3NAME	count	%	count	%	count	%	count	%	count	%	
	8.1.4	North Central Hardwood Forests	2072	13.6%	598	2.3%			1420	7.4%	171	3.1%	
	8.1.5	Driftless Area	1198	7.9%	0	0.0%			2130	11.1%	653	11.9%	
	8.1.6	Southern Michigan/Northern Indiana Drift Plains	437	2.9%	39	0.2%			895	4.7%	107	1.9%	
	8.1.7	Northeastern Coastal Zone	54	0.4%	1322	5.1%			169	0.9%	2	0.0%	
	8.1.8	Acadian Plains and Hills	51	0.3%	2848	11.1%			67	0.4%			
	8.1.10	Erie Drift Plain	129	0.8%	423	1.6%			355	1.9%	64	1.2%	
8.2	8.2.1	Southeastern Wisconsin Till Plains	333	2.2%	14	0.1%			591	3.1%	86	1.6%	
	8.2.2	Huron/Erie Lake Plains	214	1.4%	7	0.0%	4	0.1%	444	2.3%	44	0.8%	
	8.2.3	Central Corn Belt Plains	50	0.3%					167	0.9%	67	1.2%	
	8.2.4	Eastern Corn Belt Plains	278	1.8%					612	3.2%	200	3.6%	
8.3	8.3.1	Northern Piedmont	15	0.1%	10	0.0%			117	0.6%	33	0.6%	 
	8.3.2	Interior River Valleys and Hills	100	0.7%			156	2.3%	1168	6.1%	393	7.1%	
	8.3.3	Interior Plateau	126	0.8%	1	0.0%	718	10.6%	834	4.4%	572	10.4%	
	8.3.4	Piedmont	13	0.1%	29	0.1%	1071	15.8%	344	1.8%	182	3.3%	
	8.3.5	Southeastern Plains	32	0.2%	12	0.0%	763	11.3%	496	2.6%	211	3.8%	
	8.3.6	Mississippi Valley Loess Plains	2	0.0%			524	7.8%	311	1.6%	166	3.0%	
	8.3.7	South Central Plains	11	0.1%			1437	21.3%	391	2.0%	103	1.9%	
	8.3.8	East Central Texas Plains	5	0.0%			258	3.8%	58	0.3%			
8.4	8.4.1	Ridge and Valley	367	2.4%	1269	4.9%	143	2.1%	235	1.2%	177	3.2%	
	8.4.2	Central Appalachians	742	4.9%	1032	4.0%	8	0.1%	110	0.6%	128	2.3%	
	8.4.3	Western Allegheny Plateau	189	1.2%	385	1.5%	2	0.0%	869	4.5%	655	11.9%	
	8.4.4	Blue Ridge	251	1.6%	1305	5.1%	7	0.1%	24	0.1%	21	0.4%	

NA	NA 12		Ame bass Media Asso N/S	erican swood an S=5 oc S-↓ = 0.39	east hem Media Assoc N/S =	ern lock n N=8 c N-U 0.78	winge Median Assoc N/S =	<b>d elm</b> N=10 N-↓ 0.37	Americ Med N=11 Assoc N N/S =	<b>an elm</b> lian ,S=6 I-↓,S-↓ : 0.24	slippe Median N Assoc N N/S	e <b>ry elm</b> I=11, S=8 N-U, S-↓ = 0.07		
	CODE	US_L3NAME	count	%	count	%	count	%	count	%	count	%		
	8.4.5	Ozark Highlands	33	0.2%			457	6.8%	821	4.3%	468	8.5%		
	8.4.6	Boston Mountains	21	0.1%			115	1.7%	36	0.2%	43	0.8%		
	8.4.7	Arkansas Valley	2	0.0%	<u> </u>		310	4.6%	62	0.3%	24	0.4%		
	8.4.8	Ouachita Mountains	3	0.0%			327	4.8%	28	0.1%	15	0.3%		
	8.4.9	Southwestern Appalachians	66	0.4%	302	1.2%	94	1.4%	43	0.2%	29	0.5%		
8.5	8.5.1	Middle Atlantic Coastal Plain					20	0.3%	139	0.7%	49	0.9%		
	8.5.2	Mississippi Alluvial Plain	3	0.0%			223	3.3%	579	3.0%	255	4.6%		
	8.5.3	Southern Coastal Plain	1	0.0%			24	0.4%	266	1.4%	20	0.4%		
	8.5.4	Atlantic Coastal Pine Barrens									1	0.0%		
9.2	9.2.1	Northern Glaciated Plains	13	0.1%					30	0.2%				
	9.2.2	Lake Agassiz Plain	122	0.8%	<u> </u>				139	0.7%	1	0.0%	-	
	9.2.3	Western Corn Belt Plains	300	2.0%					960	5.0%	290	5.3%		
	9.2.4	Central Irregular Plains	54	0.4%	<u> </u>		27	0.4%	1250	6.5%	172	3.1%		
9.3	9.3.1	Northwestern Glaciated Plains	4	0.0%					65	0.3%	2	0.0%		
	9.3.3	Northwestern Great Plains			<u> </u>				69	0.4%	-		-	
	9.3.4	Nebraska Sand Hills							1	0.0%				
9.4	9.4.1	High Plains							9	0.0%				
	9.4.2	Central Great Plains							230	1.2%	16	0.3%		
	9.4.3	Southwestern Tablelands							10	0.1%	0	0.0%		
	9.4.4	Flint Hills	5	0.0%					129	0.7%	13	0.2%		
	9.4.5	Cross Timbers					47	0.7%	36	0.2%	5	0.1%		
	9.4.6	Edwards Plateau												

			Ame bass Media Asso N/S	erican swood an S=5 oc S-↓ = 0.39	east hem Mediar Assoc N/S =	ern lock n N=8 c N-U : 0.78	winge Median Assoc N/S =	<b>d elm</b> N=10 N-↓ 0.37	Americ Med N=11 Assoc N N/S =	<b>an elm</b> lian ,S=6 I-↓,S-↓ 0.24	slippe Median N Assoc N N/S	e <b>ry elm</b> I=11, S=8 N-U, S-↓ = 0.07		
L2	CODE	US_L3NAME	count	%	count	%	count	%	count	%	count	%		
	9.4.7	Texas Blackland Prairies					5	0.1%	3	0.0%				
9.5	9.5.1	Western Gulf Coastal Plain					20	0.3%	29	0.2%	6	0.1%		
9.6	9.6.1	Southern Texas Plains												
10.1	10.1.2	Columbia Plateau												
1	10.1.3	Northern Basin and Range												
1	10.1.4	Wyoming Basin											-	
1	10.1.5	Central Basin and Range												
1	10.1.6	Colorado Plateaus											-	
	10.1.7	Arizona/New Mexico Plateau												
1	10.1.8	Snake River Plain												
10.2	10.2.1	Mojave Basin and Range												
	10.2.2	Sonoran Basin and Range											-	
	10.2.10	Chihuahuan Deserts												
11.1	11.1.1	Southern and Central California Chaparral and Oak Woodlands												
1	11.1.2	Central California Valley												
1	11.1.3	Southern California Mountains												
12.1	12.1.1	Madrean Archipelago												
13.1	13.1.1	Arizona/New Mexico Mountains												
15.4	15.4.1	Southern Florida Coastal Plain												
		Total Tree Count	15225		25676		6760		19107		5497			

#### May 2023

#### Draft – Do Not Quote or Cite

### **APPENDIX 6.A**

## 2

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3

### DERIVATION OF THE ECOREGION AIR QUALITY METRICS (EAQM)

4 In order to better understand the relationship between past and present air quality 5 concentrations and nitrogen (N) and sulfur (S) deposition in various downwind locations of 6 significance, the EPA conducted HYSPLIT air parcel trajectory modeling to identify the 7 meteorological patterns that determine the transport of pollutant material from source to receptor. 8 Using actual air quality monitoring sites as forward trajectory starting points, the EPA was able 9 to estimate the potential regions of influence for the 84 Ecoregion III areas. After identifying the 10 upwind geographic areas from which emissions potentially contribute to N and S deposition in 11 the ecoregion, the EPA analyzed air quality design values within each ecoregion's zone of 12 influence to estimate an Ecoregion Air Quality Metric (EAQM). EAQM values were estimated 13 for each ecoregion and for four separate pollutants: NO<sub>2</sub>, SO<sub>2</sub>, and PM<sub>2.5</sub> and are intended to 14 provide a perspective of air quality levels in the upwind regions that potentially contribute to 15 downwind deposition levels. For pollutants with multiple forms of the standard, the EPA 16 estimated EAQM values for each form of the standard. This Appendix describes the 17 methodology used to calculate the air parcel trajectories that led to the zones of influence 18 identification, as well as the methodologies used to estimate the EAQM values for each 19 ecoregion/pollutant pair using historical air quality design value (DV) data.

#### 20 6A.1. HYSPLIT TRAJECTORY METHODOLOGY:

21 The Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model<sup>1</sup> is 22 commonly used to compute simple air parcel trajectories using historical meteorological data. 23 HYSPLIT simulates the trajectory of air parcels as they are transported through the atmosphere 24 for a given set of meteorological conditions. One common application of HYSPLIT is to apply 25 the model in a forward-trajectory mode to evaluate the transport of hypothetical emissions 26 releases from a specific origin. When trajectories are calculated over a large number of time 27 periods with representative meteorological conditions, one can develop a potential zone of 28 influence, or "footprint", for any emissions from this location. In this exercise, HYSPLIT was 29 used to estimate the frequency at which air transport patterns indicated that air pollutant

<sup>&</sup>lt;sup>1</sup> Stein, A.F., Draxler, R.R, Rolph, G.D., Stunder, B.J.B., Cohen, M.D., and Ngan, F., (2015). NOAA's HYSPLIT atmospheric transport and dispersion modeling system, Bull. Amer. Meteor. Soc., 96, 2059-2077, http://dx.doi.org/10.1175/BAMS-D-14-00110.1.

1 concentrations at individual monitoring sites plausibly could have contributed to deposition

- 2 within an ecoregion. The EPA was interested in assessing the air quality DVs for multiple
- 3 pollutants (and multiple forms of the standard, where relevant) in all areas that potentially
- 4 contribute to a downwind ecoregion. As explained in more detail below, multiple HYSPLIT
- 5 trajectories were generated and analyzed to determine a potential zone of influence for each
- 6 region, and then the all of the valid DVs from monitors within that area were assessed to
- 7 generate a composite "ecoregion air quality metric" (EAQM) for multiple ecoregion-pollutant
- 8 pairs.

9 The analysis used 48-hour forward trajectories with an initial plume height of 500 m and 10 a single year (2016) of meteorological data from the 32-km resolution North American Regional Reanalysis (NARR-32)<sup>2</sup>. While no single year can be considered truly representative of all 11 12 possible wind trajectories and their frequency at any given location, we note that 2016 marked 13 the transition from a strongly positive Oceanic Niño Index to a weakly negative one by the end 14 of the year. Trajectories were calculated for each monitoring site with a valid DV in the 2000-15 2018 time period. The set of sites differed by pollutant and the specific form of the standard for 16 that pollutant. In all, 568,398 individual trajectories were generated. Each 48-hour trajectory was 17 divided into 288 sequential segments corresponding to 10 minutes of the trajectory length to help ensure that we did not miss an impacting trajectory. Using geospatial tools, the EPA assessed 18 19 whether a trajectory segment from an individual monitoring site was located in an ecoregion. If 20 so, this was counted as a "hit". The analysis evaluated the frequency of "hits" for each 21 monitoring site. If more than 1% of the total hits for an ecoregion could be tracked back to a 22 monitoring site, then that site was considered to be potentially representative of air quality 23 concentrations that lead to the deposition estimated in that ecoregion. Figure 6A-1 depicts the 24 outcome for one ecoregion pollutant pair. For this ecoregion in central Kentucky, given the prevailing winds, the trajectory analysis indicates that PM<sub>2.5</sub> data from sites within the ecoregion 25 26 itself, along with 22 other sites in surrounding upwind areas (e.g., Southwest IN, Central TN) 27 may be representative of air quality levels that contribute to N and S deposition within the 28 ecoregion given the analysis parameters.

<sup>&</sup>lt;sup>2</sup> National Centers for Environmental Prediction/National Weather Service/NOAA/U.S. Department of Commerce. 2005, updated monthly. NCEP North American Regional Reanalysis (NARR). Research Data Archive at the National Center for Atmospheric Research, Computational and Information Systems Laboratory. https://rda.ucar.edu/datasets/ds608.0/. Accessed 25 May 2017.

# 6A.2. ESTIMATION OF ECOREGION AIR QUALITY METRICS (EAQMS)

After the trajectories were generated and sets of air quality monitoring sites potentially within the zone of influence were identified for each ecoregion-pollutant pair, the EPA then assessed the DVs at the sites within the contributing zone for each ecoregion-pollutant pair. These EAQMs were generated to enable an assessment of the relationship between air quality levels in the upwind contributing region to the deposition levels within the ecoregion. For each pollutant, two types of EAQMs were derived for each ecoregion based on the pollutant DVs for that ecoregion's contributing monitors:

- EAQM-max: the highest DV from any monitor within the zone of influence, and
- EAQM-weighted: a weighted average DV where each monitor's value is weighted by the
  percentage of the ecoregion's HYSPLIT hits.
- 13 Both versions of EAQMs have value. EAQM-max represents the highest DV within the 14 upwind region potentially contributing to deposition in an ecoregion, and as such it enables one 15 to determine a relationship between deposition levels (and associated adverse effects) and worst-16 case air quality that is associated with that level of deposition. Given that EAQM-weighted 17 considers the relative contributions from different upwind directions, it is presumed to represent 18 the general-case upwind air quality that is associated with downwind deposition. Both types of 19 EAQMs have inherent uncertainties related to the trajectories themselves, the methodology used 20 to link upwind regions to downwind receptors (e.g., the 1% hit assumption), and the density of
- 21 the existing monitoring network.
- Both types of EAQMs were generated for each of the 84 Ecoregion III areas for four separate combinations of pollutant and averaging time:
- SO<sub>2</sub>: annual 2<sup>nd</sup> high of individual 3-hour averages, for a single year
- SO<sub>2</sub>: annual average of 24-hour averages, for a single year
- NO<sub>2</sub>: annual average of hourly data, for a single year
- PM<sub>2.5</sub>: annual average of hourly data, averaged over 3-year periods
- 28 For the three combinations that are based on data averaged over three years, EAQMs
- 29 were generated for the following periods: 2001-2003, 2006-2008, 2010-2012, 2014-2016, and
- 30 2018-2020. For the four combinations that are based on a single year of data, EAQMs were
- 31 calculated for each of the individual 15 years between 2001 and 2020 within the identified 3-year
- 32 periods (e.g., 2001, 2002, 2003, 2006, 2007, 2008, 2010, ..., 2020). Tables 6A-1 and 6A-2 show
- example EAQM outputs for a three-year metric and a single-year metric. In Table 6A-1, which
- 34 displays the EAQM-weighted annual average PM<sub>2.5</sub> data, it can be noted that most historical
- 35 EAQM-weighted values have been below the current secondary  $PM_{2.5}$  standard of 15  $\mu$ g/m<sup>3</sup> and

- 1 that a downward trend has been observed over the past two decades. In Table 6A-2, which
- 2 displays the EAQM-max data for the annual 2<sup>nd</sup> high 3-hour SO<sub>2</sub> average (i.e., the current
- 3 secondary SO<sub>2</sub> NAAQS), it can be seen that even the worst-case maximum DVs for SO<sub>2</sub> are all
- 4 well below the 500 ppb (0.5 ppm) standard across these years and that an improving trend has
- 5 been observed between 2001 and 2020. Table 6A-3 is provided to show a sample difference
- 6 between EAQM-max and EAQM-weighted for an example case (annual average PM<sub>2.5</sub>) and
- 7 suggests, as expected, that there can be significant differences between the two types of EAQM
- 8 in some situations.
- 9 As discussed further in Chapter 6, this analytical work culminates in a series of plots 10 which display how the upwind EAQMs are related to median S and N deposition values (for
- 11 each 3-year period) within the ecoregions.
- 12



 Figure 6A-1. Map of PM<sub>2.5</sub> monitoring sites of influence (red circles) determined in the trajectory analysis to impact Ecoregion 8.3.3 (purple shaded region).
 Other PM monitoring sites determined not to impact the ecoregion are shown as gray circles.

#### 1 Table 6A-1. Example EAQM output table for a three-year average metric. These data are 2 3 the EAQM-weighted for the annual average of hourly PM2.5 data, averaged over 3-year periods ( $\mu g/m^3$ ).

	2001-	2006-	2010-	2014-	2018-
Ecoregion	2003	2008	2012	2016	2020
5.2.1	8.7	8.0	7.4	6.1	5.6
5.2.2	8.6	8.4	8.1	5.9	5.6
5.3.1	10.4	8.2	7.1	5.9	5.4
5.3.3	13.2	13.1	10.7	9.1	7.9
6.2.10	9.4	8.9	8.9	8.0	6.0
6.2.11	8.8	8.0	6.6	7.2	10.5
6.2.12	13.2	12.0	10.3	10.0	11.4
6.2.13	10.6	9.4	7.8	7.4	6.8
6.2.14	7.3	7.9	6.3	6.3	6.7
6.2.15	8.9	9.1	8.7	8.6	8.6
6.2.3	9.4	9.8	8.5	8.5	9.2
6.2.4	9.4	9.7	9.2	8.8	8.9
6.2.5	8.8	8.9	7.1	6.3	7.7
6.2.7	9.0	8.9	7.1	7.3	10.0
6.2.8	9.4	9.3	7.5	7.9	11.6
6.2.9	8.6	8.8	7.7	7.9	10.4
7.1.7	9.3	9.1	7.0	6.3	7.6
7.1.8	8.7	8.1	6.8	6.8	9.0
7.1.9	8.4	8.1	7.0	6.9	9.3
8.1.1	12.0	8.6	8.6	6.9	6.3
8.1.10	15.4	13.4	11.3	9.7	8.0
8.1.3	12.4	9.3	8.2	6.6	6.3
8.1.4	10.2	9.8	8.7	6.8	6.7
8.1.5	11.4	11.1	9.9	7.9	7.8
8.1.6	13.6	11.3	9.5	9.0	8.0
8.1.7	13.1	10.6	8.4	7.0	6.5
8.1.8	11.0	8.5	7.3	6.2	5.1
8.2.1	12.5	12.4	10.1	8.3	8.1
8.2.2	14.7	12.4	9.8	9.2	8.0
8.2.3	13.9	12.3	11.1	9.3	8.8
8.2.4	15.5	13.9	11.9	9.5	8.8
8.3.1	14.6	13.0	10.6	9.1	7.6
8.3.2	14.9	13.1	11.3	9.7	8.7
8.3.3	14.0	13.4	11.2	8.9	8.0
8.3.4	14.1	13.5	9.9	8.8	7.9
8.3.5	13.0	13.0	11.2	8.7	8.4
8.3.6	12.5	11.7	9.9	8.3	8.3
8.3.7	11.8	11.4	10.3	8.7	8.8
8.3.8	11.8	11.5	10.3	9.1	9.1
8.4.1	15.8	12.7	10.4	8.8	7.0
8.4.2	13.8	13.6	10.4	8.8	7.1
8.4.3	15.1	14.4	11.2	9.1	7.9
8.4.4	13.6	12.4	9.7	8.7	7.0
8.4.5	12.9	11.4	10.5	8.6	7.9
8.4.6	12.4	11.8	10.7	8.7	8.5
8.4.7	12.3	11.8	10.7	8.8	8.7
8.4.8	12.4	11.9	10.9	8.9	8.8

	2001-	2006-	2010-	2014-	2018-
Ecoregion	2003	2008	2012	2016	2020
8.4.9	14.6	13.8	11.2	9.0	7.9
8.5.1	11.2	11.0	8.7	7.4	6.0
8.5.2	12.2	11.3	9.8	8.2	8.1
8.5.3	9.6	8.6	9.8	6.8	7.3
8.5.4	14.5	12.6	9.9	9.2	7.9
9.2.1	7.9	8.0	8.0	5.4	5.5
9.2.2	8.0	8.2	8.1	5.5	5.6
9.2.3	10.9	9.8	9.4	7.7	7.2
9.2.4	11.8	10.6	9.9	8.1	8.0
9.3.1	8.1	8.3	8.1	6.6	6.3
9.3.3	7.8	7.2	6.9	5.5	5.1
9.3.4	7.9	7.2	6.1	5.5	6.1
9.4.1	7.8	7.3	6.5	6.3	6.9
9.4.2	10.1	9.4	8.9	8.2	8.6
9.4.3	7.2	7.0	6.4	6.3	6.7
9.4.4	11.0	10.0	9.5	8.2	8.6
9.4.5	11.5	10.8	10.1	8.9	9.0
9.4.6	11.0	10.5	9.5	8.9	8.8
9.4.7	12.2	11.2	10.2	9.0	9.2
9.5.1	11.1	11.2	10.0	8.9	9.0
9.6.1	10.9	10.8	9.8	9.2	9.0
10.1.2	8.6	9.1	7.6	7.3	9.2
10.1.3	9.5	9.0	7.8	8.0	10.5
10.1.4	10.6	8.9	7.4	6.0	5.7
10.1.5	12.6	11.2	9.1	8.8	8.2
10.1.6	9.2	9.2	6.9	7.1	6.5
10.1.7	7.4	8.0	6.5	7.1	7.0
10.1.8	9.5	9.1	8.2	8.1	8.5
10.2.1	16.7	13.9	10.7	10.2	10.1
10.2.2	14.4	10.9	8.8	7.7	8.3
10.2.4	8.1	8.6	7.8	7.2	7.2
11.1.1	14.5	11.4	9.1	8.1	9.0
11.1.2	14.1	12.5	10.6	10.1	12.0
11.1.3	18.6	13.7	10.4	9.6	9.4
12.1.1	8.8	8.7	7.5	7.1	7.3
13.1.1	8.4	8.6	7.0	7.1	7.4
15.4.1	8.4	7.8	7.4	6.4	7.1

1	
2	

Table 6A-2. Example EAQM output table for a single-year metric. These data are theEAQM-max for the annual 2<sup>nd</sup> high of individual 3-hour SO2 averages (ppb).

Ecoregion	2001	2002	2003	2006	2007	2008	2010	2011	2012	2014	2015	2016	2018	2019	2020
5.2.1	138	139	128	186	182	229	145	199	176	153	126	106	83	65	62
5.2.2	96	95	107	55	82	106	55	63	76	47	49	66	41	65	45
5.3.1	96	51	69	42	45	204	221	193	59	64	14	12	80	77	75
5.3.3	249	319	264	231	188	152	155	332	117	116	111	81	78	70	51
6.2.10	112	246	200	62	91	60	118	40	55	57	68	38	34	33	57
6.2.11	84	78	78	74	55	48	19	20	14	14	13	9	17	13	10
6.2.12	84	78	78	74	55	48	12	15	44	19	13	9	17	13	10
6.2.13	102	246	200	62	91	42	118	35	55	50	68	38	34	33	57
6.2.14	219	246	200	162	174	169	538	116	216	178	147	257	132	150	57
6.2.15	112	246	200	74	91	57	118	39	55	57	68	27	82	65	57
6.2.3	112	91	99	62	64	57	65	39	55	57	68	27	82	65	54
6.2.4	112	91	99	62	64	57	118	39	55	57	68	27	82	65	54
6.2.5	28	41	23	20	24	24	19	20	14	10	13	9	82	65	54
6.2.7	34	44	23	20	24	48	19	20	14	14	13	9	82	65	54
6.2.8	34	44	23	20	24	48	19	20	14	14	13	9	82	65	54
6.2.9	112	246	200	62	91	48	118	35	55	50	68	27	34	33	57
7.1.7	28	41	23	18	24	24	19	20	14	4	13	3	82	65	54
7.1.8	34	44	23	20	24	48	19	20	14	14	13	9	82	65	54
7.1.9	34	44	23	18	24	24	19	20	14	11	8	9	82	65	54
8.1.1	245	212	264	200	158	125	118	132	169	116	131	361	83	77	75
8.1.10	121	123	174	231	178	126	118	132	169	116	131	361	83	49	56
8.1.3	245	273	264	200	129	125	93	102	60	116	85	34	50	47	35
8.1.4	138	139	128	186	182	229	145	199	176	153	126	106	83	65	62
8.1.5	100	103	240	236	312	229	202	199	219	182	124	38	26	65	45
8.1.6	107	85	122	109	193	234	149	153	89	92	101	96	83	58	62
8.1.7	135	105	108	126	134	204	221	193	59	64	25	12	12	20	19
8.1.8	135	105	108	126	134	204	221	193	59	27	25	12	80	77	75
8.2.1	331	179	240	236	312	234	202	199	219	182	126	106	83	58	62
8.2.2	113	118	176	109	106	123	121	132	169	76	131	361	83	58	58
8.2.3	331	179	240	236	312	311	202	199	219	182	124	96	78	58	56
8.2.4	158	178	146	165	193	234	149	153	134	142	101	96	91	151	60
8.3.1	91	91	87	75	123	77	61	332	117	77	69	46	182	129	51
8.3.2	254	226	291	235	318	311	202	199	219	182	124	96	361	386	357
8.3.3	182	225	196	165	164	162	107	153	134	142	101	96	361	386	59
8.3.4	170	235	145	168	194	188	158	138	126	46	41	38	191	87	184
8.3.5	114	123	137	149	237	78	235	293	178	77	67	63	67	60	54
8.3.6	254	210	238	121	237	311	235	293	181	77	67	63	361	386	357
8.3.7	142	105	236	151	68	98	65	99	94	44	52	57	73	92	70
8.3.8	142	105	236	151	73	84	65	99	94	44	52	57	73	92	70
8.4.1	170	134	145	168	194	188	158	138	126	77	111	81	191	129	184
8.4.2	197	244	175	168	194	188	158	332	126	77	111	81	191	129	184
8.4.3	249	319	237	231	188	203	155	332	117	108	111	81	78	70	55
8.4.4	170	235	145	168	194	188	158	138	126	46	41	38	191	87	184
8.4.5	254	226	238	224	318	311	140	146	181	110	117	38	361	386	357
8.4.6	254	226	238	151	127	143	140	146	181	110	117	57	73	92	70

Ecoregion	2001	2002	2003	2006	2007	2008	2010	2011	2012	2014	2015	2016	2018	2019	2020
8.4.7	142	210	236	151	127	143	94	146	144	110	117	57	73	92	70
8.4.8	142	105	236	151	87	92	94	99	94	44	52	57	73	92	70
8.4.9	170	235	145	168	194	188	107	77	126	115	53	38	191	60	31
8.5.1	93	141	139	119	83	77	80	58	82	60	34	35	61	50	47
8.5.2	254	226	238	151	237	98	235	293	181	77	67	63	361	386	357
8.5.3	135	123	137	149	120	135	160	66	110	60	75	57	35	34	54
8.5.4	91	141	139	256	141	77	42	91	59	64	21	46	11	10	9
9.2.1	112	95	107	57	82	106	65	63	76	57	49	66	173	133	137
9.2.2	96	95	107	57	82	106	55	63	76	57	49	66	41	65	45
9.2.3	126	103	113	121	182	143	94	199	176	153	124	66	41	65	45
9.2.4	128	94	87	114	127	143	94	146	144	110	117	66	41	92	70
9.3.1	112	95	107	62	91	60	118	63	55	57	68	27	34	33	57
9.3.3	112	246	200	62	91	60	118	63	55	57	68	38	34	33	57
9.3.4	112	246	200	62	91	60	118	63	55	57	68	46	173	133	137
9.4.1	219	246	200	162	174	169	538	116	216	178	147	257	283	150	137
9.4.2	128	86	87	99	73	84	55	146	76	42	46	46	283	133	137
9.4.3	219	179	165	162	174	169	538	116	216	178	147	257	283	150	137
9.4.4	128	86	87	114	127	143	94	146	144	110	117	66	283	133	137
9.4.5	142	86	93	99	73	84	55	99	47	44	34	38	283	92	87
9.4.6	142	105	236	151	73	84	65	66	47	44	34	20	283	92	87
9.4.7	142	105	236	151	73	84	65	99	94	44	52	57	73	92	70
9.5.1	142	105	236	151	237	98	235	293	178	77	52	63	73	92	70
9.6.1	142	105	236	151	73	84	65	66	47	44	52	57	283	92	87
10.1.2	112	91	87	62	51	48	19	20	55	35	68	27	82	65	54
10.1.3	102	246	200	74	91	48	118	35	55	50	68	27	34	33	57
10.1.4	102	246	200	62	91	57	118	39	55	57	68	38	34	33	57
10.1.5	84	78	78	74	55	48	26	30	55	35	68	27	34	13	42
10.1.6	102	66	200	99	98	82	118	84	71	143	147	257	132	88	57
10.1.7	219	179	165	162	174	169	538	116	216	178	147	257	132	150	46
10.1.8	102	246	200	62	91	48	118	35	55	50	68	27	34	33	57
10.2.1	84	78	78	74	55	37	26	30	44	19	13	11	17	13	10
10.2.2	219	179	165	162	174	169	538	116	216	178	147	257	132	150	46
10.2.4	219	179	165	162	174	169	538	116	216	178	147	257	283	150	137
11.1.1	84	78	78	74	55	48	26	30	44	19	13	11	17	13	10
11.1.2	84	78	78	74	55	48	12	15	44	19	13	9	17	13	10
11.1.3	84	78	78	74	55	37	26	30	44	19	13	11	6	13	10
12.1.1	219	179	165	162	174	169	538	116	216	178	147	257	132	150	46
13.1.1	219	179	165	162	174	169	538	116	216	178	147	257	132	150	46
15.4.1	135	119	129	93	120	135	96	66	110	51	75	57	35	16	31

# Table 6A-3. Example table showing differences between an EAQM-max and EAQM weighted values. These sample data are for the annual average of hourly PM<sub>2.5</sub> metric, averaged over 3-year periods (μg/m<sup>3</sup>). The data are EAQM-max" minus EAQM-weighted.

Ecoregion	2003	2008	2012	2016	2020
5.2.1	3.3	3.0	2.3	2.6	2.7
5.2.2	3.4	2.6	3.4	2.8	2.7
5.3.1	1.4	2.8	2.5	2.2	2.1
5.3.3	2.7	5.2	4.1	3.7	3.2
6.2.10	4.6	2.8	2.3	4.4	4.9
6.2.11	4.6	5.4	3.7	3.2	5.8
6.2.12	8.6	9.5	8.9	8.4	6.2
6.2.13	3.4	1.7	1.4	2.9	2.9
6.2.14	3.6	3.2	4.0	2.9	3.2
6.2.15	6.2	4.3	2.8	3.8	7.7
6.2.3	6.6	3.9	3.0	3.9	4.1
6.2.4	6.6	4.0	2.3	3.6	4.4
6.2.5	2.3	2.8	1.6	2.5	4.2
6.2.7	4.4	4.5	3.2	3.1	6.3
6.2.8	5.7	4.1	4.0	7.1	4.7
6.2.9	6.5	4.6	3.8	7.1	5.9
7.1.7	1.8	2.5	1.8	2.5	3.1
7.1.8	4.7	5.3	3.5	2.7	4.9
7.1.9	5.0	3.5	3.3	2.6	4.6
8.1.1	1.9	2.2	4.6	4.5	1.2
8.1.10	2.9	1.7	1.9	2.4	1.3
8.1.3	1.5	1.5	1.4	1.6	0.9
8.1.4	1.8	2.3	1.7	1.9	1.6
8.1.5	1.6	2.8	2.3	1.5	1.0
8.1.6	1.6	0.9	2.7	1.4	1.6
8.1.7	4.4	3.7	1.5	3.2	2.0
8.1.8	3.2	2.5	2.3	1.9	2.5
8.2.1	1.7	2.1	2.1	1.1	2.5
8.2.2	4.8	3.0	1.7	2.1	2.9
8.2.3	2.2	2.3	1.1	0.8	1.8
8.2.4	2.6	1.8	1.5	1.9	2.8
8.3.1	2.4	1.9	1.7	3.7	1.7
8.3.2	4.2	2.6	1.1	0.4	1.8
8.3.3	2.9	1.9	2.0	1.5	2.1
8.3.4	1.5	1.7	2.4	1.6	1.6
8.3.5	0.6	1.6	1.9	1.4	0.7
8.3.6	1.5	2.3	2.0	1.5	1.3
8.3.7	2.4	3.8	1.8	2.1	1.8
8.3.8	2.4	3.7	1.8	1.7	1.5
8.4.1	0.0	0.0	1.7	1.6	1.1
8.4.2	3.3	1.8	2.1	1.9	1.6
8.4.3	2.1	1.0	3.6	3.7	3.2
8.4.4	2.8	2.7	2.4	1.7	2.5

Ecoregion	2003	2008	2012	2016	2020
8.4.5	3.8	1.8	1.4	1.5	2.4
8.4.6	1.7	0.8	1.4	2.1	2.1
8.4.7	1.8	3.4	1.4	2.0	1.9
8.4.8	1.8	3.3	1.2	1.9	1.8
8.4.9	3.4	3.5	1.8	2.2	2.1
8.5.1	0.3	1.7	0.7	0.7	1.0
8.5.2	1.9	1.6	2.1	1.6	1.5
8.5.3	4.2	4.1	3.3	2.5	1.8
8.5.4	1.7	1.9	3.2	3.6	1.5
9.2.1	2.8	2.8	3.5	3.3	2.7
9.2.2	2.7	2.6	3.4	3.2	2.6
9.2.3	3.0	2.2	2.1	1.8	2.4
9.2.4	2.1	1.4	1.6	1.4	2.0
9.3.1	7.9	5.4	3.4	5.8	7.0
9.3.3	5.0	4.0	4.3	6.9	4.4
9.3.4	4.9	3.9	3.2	3.7	3.8
9.4.1	3.7	4.6	4.0	3.1	3.0
9.4.2	3.8	2.4	1.9	1.7	1.4
9.4.3	4.3	4.9	4.1	3.1	3.2
9.4.4	2.9	2.0	2.0	1.4	1.4
9.4.5	2.7	4.4	2.0	1.9	1.6
9.4.6	3.2	4.7	2.6	1.9	1.8
9.4.7	2.0	4.0	1.9	1.8	1.4
9.5.1	3.1	4.1	2.1	1.9	1.6
9.6.1	3.3	4.4	2.3	1.6	1.6
10.1.2	4.8	4.6	3.9	4.6	7.1
10.1.3	5.6	4.4	3.7	7.0	5.8
10.1.4	3.4	2.2	3.8	1.8	2.6
10.1.5	12.6	10.3	6.9	9.6	8.4
10.1.6	4.8	1.9	6.7	5.5	3.2
10.1.7	8.4	5.0	7.1	5.5	5.8
10.1.8	5.6	4.3	3.3	6.9	7.8
10.2.1	11.1	7.6	5.3	8.2	7.6
10.2.2	13.4	9.1	6.4	6.8	4.5
10.2.4	3.6	5.1	2.7	2.9	5.6
11.1.1	13.4	8.6	10.1	6.4	4.0
11.1.2	7.7	9.0	8.6	8.3	5.6
11.1.3	9.2	7.8	5.2	8.8	4.8
12.1.1	2.9	5.0	3.0	2.4	5.5
13.1.1	7.4	5.1	6.6	5.5	5.4
15.4.1	2.6	1.7	5.7	2.0	2.0

# Table 6A-4. Table of the median TDEP S deposition estimated for each Ecoregion III areacompared to the median TDEP deposition estimate for each of the water bodylocations used in Ecoregion III areas in the aquatic critical load analysis inChapter 5

	Median S				
	Deposition at				
	Aquatic CL	Median S		Damaant	
Pegion	Sites from	from TDEP	Difference	Difference	Voar
8 1 1	8.04		2 0/	30.02%	2001-2003
811	6.50	8.82	2.34	30.32 %	2001-2003
533	3.22	1 09	0.87	23 91%	2000-2000
533	5.83	7.03	1 /1	23.3170	2014-2010
8 1 1	3.05	1.24	0.78	21.3270	2010-2012
0.1.1 Q 1 1	0.20 0.00	2.04	0.78	10.64%	2010-2012
533	15 73	19.09	2.40	13.04 /0	2014-2010
0.0.0	1.7.5	164	2.30	13.95 /0	2001-2003
0.1.1	1.44	1.04	0.20	12.09%	2010-2020
0.3.3	3.07	4.10	0.49	12.30%	2014-2010
0.0.0	13.37	10.00	1.00		2006-2008
8.3.3	5.58	0.24	0.00	11.10%	2010-2012
8.3.3	9.84	10.96	1.12	10.74%	2006-2008
5.3.3	2.17	2.40	0.23	10.04%	2018-2020
8.4.1	1.94	2.14	0.20	9.61%	2018-2020
8.3.5	2.41	2.63	0.23	8.94%	2018-2020
5.2.1	4.01	4.29	0.28	6.73%	2001-2003
5.2.1	3.10	3.24	0.14	4.41%	2006-2008
5.2.1	2.34	2.44	0.10	4.09%	2010-2012
8.1.7	2.32	2.40	0.07	3.18%	2014-2016
8.3.3	13.11	13.52	0.41	3.10%	2001-2003
8.1.7	9.29	9.57	0.28	3.00%	2001-2003
8.1.7	3.71	3.82	0.11	2.97%	2010-2012
8.4.2	4.00	4.12	0.11	2.83%	2014-2016
8.1.3	2.71	2.79	0.08	2.73%	2014-2016
8.3.7	4.58	4.70	0.12	2.66%	2014-2016
8.1.3	4.69	4.81	0.12	2.52%	2010-2012
8.3.4	4.24	4.34	0.10	2.25%	2010-2012
8.1.3	11.69	11.92	0.24	2.02%	2001-2003
8.1.7	8.28	8.42	0.14	1.71%	2006-2008
5.2.1	1.31	1.33	0.01	0.99%	2018-2020
8.3.5	3.44	3.48	0.03	0.97%	2014-2016
5.2.1	1.88	1.89	0.02	0.81%	2014-2016
8.4.4	4.41	4.41	-0.01	-0.11%	2010-2012
8.3.1	3.34	3.32	-0.01	-0.35%	2014-2016

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2 3

	Median S				
	Deposition at				
	Aquatic CL	Median S		Parcont	
Region	TDEP	from TDEP	Difference	Difference	Year
8.3.3	2.74	2.73	-0.01	-0.37%	2018-2020
841	14 18	14 10	-0.08	-0.60%	2001-2003
8.4.1	11.93	11.86	-0.08	-0.66%	2006-2008
8.4.4	11.29	11.12	-0.17	-1.50%	2001-2003
8.3.1	15.18	14.94	-0.24	-1.61%	2001-2003
8.4.4	2.65	2.61	-0.05	-1.81%	2014-2016
8.1.7	1.91	1.87	-0.03	-1.85%	2018-2020
8.1.3	10.45	10.24	-0.20	-1.98%	2006-2008
8.4.9	5.59	5.47	-0.12	-2.20%	2010-2012
8.3.7	5.03	4.91	-0.11	-2.29%	2010-2012
8.4.5	3.27	3.19	-0.08	-2.45%	2014-2016
8.1.3	1.73	1.68	-0.04	-2.60%	2018-2020
8.4.5	2.66	2.59	-0.07	-2.69%	2018-2020
8.3.1	12.94	12.58	-0.36	-2.82%	2006-2008
8.4.2	7.25	7.05	-0.21	-2.91%	2010-2012
8.4.4	9.58	9.26	-0.32	-3.39%	2006-2008
5.3.1	3.12	3.01	-0.11	-3.49%	2010-2012
8.3.4	2.72	2.62	-0.10	-3.72%	2014-2016
8.3.1	2.21	2.12	-0.08	-3.87%	2018-2020
8.4.5	4.87	4.65	-0.22	-4.59%	2010-2012
8.3.4	12.26	11.71	-0.55	-4.61%	2001-2003
8.4.2	2.43	2.32	-0.11	-4.80%	2018-2020
8.4.2	17.03	16.20	-0.82	-4.96%	2001-2003
8.4.1	3.40	3.23	-0.17	-5.09%	2014-2016
8.4.2	13.98	13.28	-0.71	-5.19%	2006-2008
8.3.7	7.15	6.78	-0.36	-5.22%	2006-2008
8.4.4	2.06	1.95	-0.11	-5.33%	2018-2020
8.3.7	7.77	7.34	-0.43	-5.63%	2001-2003
8.3.4	10.14	9.58	-0.56	-5.67%	2006-2008
8.4.5	6.18	5.84	-0.35	-5.75%	2006-2008
5.3.1	6.12	5.78	-0.34	-5.79%	2006-2008
8.3.1	5.63	5.30	-0.33	-6.07%	2010-2012
8.1.4	1.48	1.39	-0.09	-6.40%	2018-2020
8.3.7	3.88	3.64	-0.24	-6.44%	2018-2020
8.3.4	2.03	1.89	-0.14	-7.22%	2018-2020
8.4.1	5.71	5.31	-0.41	-7.40%	2010-2012
8.1.4	3.72	3.42	-0.30	-8.46%	2006-2008
8.1.4	2.86	2.63	-0.23	-8.49%	2010-2012

Region	Median S Deposition at Aquatic CL Sites from TDEP	Median S Deposition from TDEP	Difference	Percent Difference	Year
8.1.4	2.19	2.01	-0.18	-8.53%	2014-2016
8.4.5	6.95	6.31	-0.64	-9.68%	2001-2003
5.3.1	1.48	1.34	-0.14	-10.06%	2018-2020
8.3.5	4.83	4.34	-0.48	-10.53%	2010-2012
5.3.1	2.23	1.99	-0.23	-11.07%	2014-2016
8.1.8	4.98	4.46	-0.52	-11.12%	2001-2003
8.4.9	2.93	2.61	-0.32	-11.45%	2018-2020
8.3.5	10.88	9.68	-1.20	-11.65%	2001-2003
5.3.1	7.29	6.46	-0.84	-12.15%	2001-2003
8.3.5	9.14	8.05	-1.09	-12.68%	2006-2008
8.1.4	5.30	4.57	-0.73	-14.70%	2001-2003
8.4.9	17.27	14.71	-2.56	-16.03%	2001-2003
8.1.8	5.42	4.61	-0.81	-16.14%	2006-2008
8.1.8	1.94	1.65	-0.29	-16.15%	2014-2016
8.1.8	1.44	1.22	-0.22	-16.49%	2018-2020
8.1.8	2.83	2.38	-0.45	-17.27%	2010-2012
8.4.9	4.17	3.46	-0.72	-18.83%	2014-2016
8.4.9	14.44	11.56	-2.89	-22.19%	2006-2008

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