Draft Scope and Methods Plan for Risk/Exposure Assessment: Secondary NAAQS Review for Oxides Of Nitrogen and Oxides of Sulfur

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EPA-452/D-08-002
March 2008

Draft Scope and Methods Plan for Risk/Exposure Assessment: Secondary NAAQS Review for Oxides Of Nitrogen and Oxides of Sulfur

Office of Air Quality Planning and Standards
US Environmental Protection Agency
Research Triangle Park, North Carolina 27711

Disclaimer

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Key Terms

Acidification: The process of increasing the acidity of a system (e.g., lake, stream, forest soil). Atmospheric deposition of acidic or acidifying compounds can acidify lakes, streams, and forest soils.

Adverse Effect: The response or component of an ecosystem that is deemed harmful in its function.

Air Quality Indicator: The substance or set of substances (e.g., $PM_{2.5}$, NO_2, SO_2) occurring in the ambient air for which the National Ambient Air Quality Standards set a standard level and monitoring occurs.

Alpine: The biogeographic zone made up of slopes above the tree line, characterized by the presence of rosette-forming herbaceous plants and low, shrubby, slow-growing woody plants.

Acid Neutralizing Capacity: A key indicator of the ability of water to neutralize the acid or acidifying inputs it receives. This ability depends largely on associated biogeophysical characteristics.

Arid Region: A land region of low rainfall, where “low” is widely accepted to be less than 250 mm precipitation per year.

Assessment Endpoint: An ecological entity and its attributes impacts to which are considered welfare effects, as defined in Clean Air Act Section 302(h), and that are analyzed in the assessment.

Base Cation Saturation: The degree to which soil cation exchange sites are occupied with base cations (e.g., Ca^{2+}, Mg^{2+}, K^+) as opposed to Al^{3+} and H^+. Base cation saturation is a measure of soil acidification, with lower values being more acidic. There is a threshold whereby soils with base saturations less than 20% (especially between 10–20%) are extremely sensitive to change.

Biologically Relevant Indicator: A physical, chemical, or biological entity/feature that demonstrates a consistent degree of response to a given level of stressor exposure and that is easily measured/quantified to make it a useful predictor of biological, environmental, or ecological risk.

Buffering Capacity: The ability of a body of water and its watershed to neutralize introduced acid.

Critical Load: A quantitative estimate of the level of exposure to one or more pollutants, below which significant harmful effects on specific sensitive elements of the environment do not occur according to present knowledge.

Denitrification: The anaerobic reduction of oxidized nitrogen (e.g., nitrate or nitrite) to gaseous nitrogen (e.g., N_2O or N_2) by denitrifying bacteria.

Dry Deposition: The removal of gases and particles from the atmosphere to surfaces in the absence of precipitation (rain, snow) or occult deposition.

Ecological Dose: The concentration of a toxicant that inhibits a microbe-mediated ecological process by designated percentage; for example, ED50 inhibits 50%.

Ecological Exposure: The exposure of a nonhuman organism to an environmental stressor.

Ecological Risk: The likelihood that adverse ecological effects may occur or are occurring as a result of exposure to one or more stressors (U.S. EPA, 1992a).

Ecological Risk Assessment: A process that evaluates the likelihood that adverse ecological effects may occur or are occurring as a result of exposure to one or more stressors (U.S. EPA, 1992a).

Ecosystem: The dynamic, complex interaction of plants, animals, and microorganisms and the nonliving environment.

Ecosystem Benefit: The value, expressed qualitatively, quantitatively, and/or in economic terms, where possible, associated with changes in ecosystem services that result either directly or indirectly in improved human health and/or welfare. Examples of ecosystem benefits that derive from improved air quality include improvements in habitats for sport fish species, the quality of drinking water and recreational areas, and the visual quality of scenic views.

Ecosystem Function: The processes and interactions that operate within an ecosystem.

Ecosystem Services: The ecological processes or functions having monetary or nonmonetary value to individuals or society at large. There are (i) supporting services, such as productivity or biodiversity maintenance; (ii) provisioning services, such as food, fiber, or fish; (iii) regulating services, such as climate regulation or carbon sequestration; and (iv) cultural services, such as tourism or spiritual and aesthetic appreciation.

Elasticity: The percentage of change in the response variable for a 1% change in the input physical or meteorological characteristic.

Eutrophication: The process by which nitrogen additions stimulate the growth of autotrophic biota, usually resulting in the depletion of dissolved oxygen.

Greenhouse Gas: Those gaseous constituents of the atmosphere, both natural and anthropogenic, that absorb and emit radiation at specific wavelengths within the spectrum of infrared radiation emitted by the earth's surface, the atmosphere, and clouds. This property causes the greenhouse effect. H_2O vapor, CO_2, N_2O, CH_4, and O_3 are the primary greenhouse gases in the earth's atmosphere. As well as CO_2, N_2O, and CH_4, the Kyoto Protocol deals with SF_6, hydrofluorocarbons, and perfluorocarbons.

Nitrogen Enrichment: The process by which a terrestrial system becomes enriched by nutrient additions to a degree that stimulates the growth of plant or other terrestrial biota, usually resulting in an increase in productivity.

Nitrogen Saturation: The condition when nitrogen inputs from atmospheric deposition and other sources exceed the biological requirements of the ecosystem.

Occult Deposition: The removal of gases and particles from the atmosphere to surfaces by fog or mist.

Semiarid Regions: Those regions of moderately low rainfall, typically 25 to 50 centimeters (10 to 20 inches) of rainfall per year, where the natural vegetation is usually short grasses and shrubs and where the predominant land use may be as rangelands.

Sensitivity: The degree to which a system is affected, either adversely or beneficially, by SO_x or NO_x pollution (e.g., from acidification or nitrogen nutrient enrichment). The impacts to natural environmental systems can be reflected in direct changes in growth or survival rates for individual species or changes at a community level reflected in shifts in measures such as species diversity.

Target Load: A policy-based metric that takes into consideration such factors as economic costs and time frame for emissions reduction. This can be lower than the critical load if a very sensitive area is to be protected in the short term, especially if deposition rates exceed critical loads.

Total Reactive Nitrogen: This includes all biologically, chemically, and radiatively active nitrogen compounds in the atmosphere and biosphere, such as NH_3, NH_4^+, NO, NO_2, HNO_3, N_2O, NO_3^-, and organic compounds (e.g., urea, amines, nucleic acids).

Valuation: The economic or noneconomic process of determining either the value of maintaining a given ecosystem type, state, or condition or the value of a change in an ecosystem, its components, or the services it provides.

Vulnerability: The degree to which a system is susceptible to, and unable to cope with, the adverse effects of NO_x and/or SO_x air pollution. Vulnerability is a function of the exposed and its sensitivity.

Welfare Effects: The effects on soils; water; crops; vegetation; man-made materials; animals; wildlife; weather; visibility; and climate as well as damage to, and deterioration of, property; hazards to transportation; and the effects on economic values and on personal comfort and well-being, whether caused by transformation, conversion, or combination with other air pollutants (Clean Air Act Section 302[h]).

Wet Deposition: The removal of gases and particles from the atmosphere to surfaces by rain or other precipitation.

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1. CONTEXT AND INTRODUCTION

1.1 Context for This Scope and Methods Plan

The U.S. Environmental Protection Agency (EPA) is currently conducting a joint review of the existing secondary (welfare-based) National Ambient Air Quality Standards (NAAQS) for nitrogen dioxide (NO_2) and sulfur dioxide (SO_2). We recognize that this is the first time that we have conducted a joint, multi-pollutant review of a secondary standard separate from the review of the primary standard. As discussed in the Integrated Review Plan (U.S. EPA, 2007a), this was done in recognition of the important linkages between ambient nitrogen and sulfur leading to deposition of ambient particles that can have significant impacts on the environment. We further recognize that a fully comprehensive assessment of such linkages and impacts is very complex and will extend beyond the time available in this review, as constrained by our court-ordered schedule. Thus, this Scope and Methods Plan is more narrowly focused on key aspects of the evolving scientific understanding to provide timely results to meet our court-ordered schedule. Our plan for the current review is to focus on the identification of sensitive ecosystems, the predominant linkages between ambient levels of nitrogen and sulfur, and the levels of deposition that create adverse effects in those ecosystems, building directly from the key findings of our Integrated Science Assessment (ISA). To the degree possible, our risk and exposure assessment will attempt to evaluate whether ecosystem damage is occurring in specific ecosystems under current ambient concentrations, and, if so, what alternative levels of ambient nitrogen and sulfur might be expected to allow various degrees of recovery of impacted systems and prevention of further damage.

In particular, given the data and time constraints of our current review, we plan to base our overall assessment on a small number of local or regional case studies where adequate data are available to enable the quantification of some of the more important linkages, focusing on the impacts of acidification and enrichment for both terrestrial and aquatic systems. Along with these case studies, we plan to conduct statistical and spatial characterizations of existing national-scale databases on air quality, nitrogen and sulfur deposition, and ecosystem characteristics to help place the results of these local or regional case studies in a broader spatial context. The combination of these case studies and the national characterizations thus forms the body of our planned overall assessment. Additional case studies and more comprehensive investigations of a broader set of effects, linkages, and indicators may be identified during this process that might reasonably form the basis for further assessment in the next 5-year review cycle, when there will be a more robust database on which to develop a more comprehensive understanding of these relationships.

1.2 Introduction

The reviews of the primary NAAQS for NO_2 and SO_2 are addressed in separate plans released during the winter of 2006–2007. The revised, secondary NAAQS review process contains four major components: an integrated review plan, a science assessment, a risk/exposure assessment, and a policy assessment/rulemaking. This Scope and Methods Plan is the first phase of the risk/exposure assessment; it will describe the scope of the analyses to be performed and the tools and methods that will be used for the joint review of the secondary NAAQS for these pollutants. In this plan, the terms NO_2 and oxides of nitrogen (NO_x) and SO_2 and oxides of sulfur (SO_x) are not interchangeable. The terms NO_x and SO_x refer to the listed Criteria Air Pollutants for which EPA has regulatory authority under Sections 108 and 109 of the Clean Air Act (CAA), and for which criteria must be developed and reviewed every 5 years. It is necessary to distinguish between the definition of "nitrogen oxides" as it appears in the enabling legislation related to the NAAQS and the definition commonly used in the air pollution research and management community. In this document, the terms "oxides of nitrogen" and "nitrogen oxides" refer to all forms of oxidized nitrogen compounds, including nitric oxide (NO), nitrogen dioxide (NO_2), and all other oxidized nitrogen-containing compounds transformed from NO and NO_2. This follows usage in the Clean Air Act Section 108(c): "Such criteria [for oxides of nitrogen] shall include a discussion of nitric and nitrous acids, nitrites, nitrates, nitrosamines, and other carcinogenic and potentially carcinogenic derivatives of oxides of nitrogen." By contrast, within the air pollution research and control community, the terms "oxides of nitrogen" and "nitrogen oxides" are restricted to refer only to the sum of NO and NO_2, and this sum is commonly abbreviated as NO_x. The category label used by this community for the sum of all forms of oxidized nitrogen compounds including those listed in Section 108(c) is NO_y.

The terms NO_2 and SO_2 refer to the specific air quality indicators (pollutant species) specified by the current standards whose concentrations are monitored to determine whether the NAAQS are being met in a given location. The ecological importance of both oxidized and reduced forms of nitrogen has been widely recognized by the scientific community. Therefore, this risk/exposure assessment will also evaluate total reactive nitrogen (which includes both oxidized and reduced forms of nitrogen) and its impacts on public welfare. It is addressed in this NAAQS review because reduced forms of nitrogen may also cause many of the effects resulting from oxides of nitrogen (e.g., deposition-influenced nitrogen enrichment).

Because NO_x, SO_x, and their associated transformation products are linked from an atmospheric chemistry perspective, as well as from an environmental effects perspective, and because of the National Research Council's (NRC's) 2004 recommendations to consider multiple pollutants in forming the scientific basis for the NAAQS, EPA has decided, for the first time since NAAQS were established in 1971, to jointly assess the science, risks, and policies relevant to protect the public welfare associated with oxides of nitrogen and oxides of sulfur. Though these interactions have been recognized historically by both the Clean Air Scientific Advisory Committee (CASAC) and EPA, and the science related to these interactions has continued to evolve and grow to the present day, providing ongoing support for considering them together.

This Scope and Methods Plan is organized to provide information consistent with EPA's 1998 *Guidelines for Ecological Risk Assessment* (U.S. EPA, 1998) that is representative of the problem formulation phase of the risk assessment. The Scope and Methods Plan is organized to provide the following:

- Background on NAAQS legislation, previous secondary reviews, an overview of ecological risk assessments, and the scope of this NAAQS review
- A conceptual model of nitrogen and sulfur cycling
- Key policy-relevant questions
- EPA's proposed schedule for the NO_x/SO_x secondary NAAQS review
- An analysis plan that includes the identification of relevant indicators of effects, a proposed approach to select areas for the risk/exposure assessment, an evaluation of data and models to assess effects, plans for characterization of exposure, and plans for characterization of ecological effects
- An assessment of alternative levels of protection under different scenarios of deposition from ambient sources.

EPA is consulting with CASAC, an independent scientific advisory committee established under the CAA, on this plan. In particular, given the context for this NAAQS review and the timing constraints for its completion, we are soliciting the advice from CASAC on how to best focus our current assessment activities to provide meaningful results to inform the regulatory portion of this review. To aid in this process, we have identified our initial priorities for each step in the process (i.e. case study areas, larger assessment areas, endpoints and indicators) for the overall risk/exposure assessment. As this review proceeds, the plan described here may be modified to reflect information received during the review process and to address advice and comments received from CASAC and the public.

2. BACKGROUND AND OVERVIEW

2.1 Legislative Requirements

Two sections of the CAA govern the establishment and revision of the NAAQS Section 108 (42 U.S.C. 7408) directs the Administrator to identify and list "air pollutants" that "in his judgment, may reasonably be anticipated to endanger public health and welfare" and whose "presence . . . in the ambient air results from numerous or diverse mobile or stationary sources" and to issue air quality criteria for those that are listed. Air quality criteria are intended to "accurately reflect the latest scientific knowledge useful in indicating the kind and extent of identifiable effects on public health or welfare which may be expected from the presence of [a] pollutant in ambient air. . . ."

Section 109 (42 U.S.C. 7409) directs the Administrator to propose and promulgate "primary" and "secondary" NAAQS for pollutants listed under Section 108. A secondary standard, as defined in Section 109(b)(2), must "specify a level of air quality the attainment and maintenance of which, in the judgment of the Administrator, based on such criteria, is required to protect the public welfare from any known or anticipated adverse effects associated with the presence of [the] pollutant in the ambient air." Welfare effects, as defined in Section 302(h) [42 U.S.C. 7602(h)], include, but are not limited to, "effects on soils, water, crops, vegetation, man-made 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." The definition of public welfare in Section 302(h) was expanded in the 1990 CAA amendments to state that the welfare effects identified should be protected from adverse effects associated with criteria air pollutants "whether caused by transformation, conversion, or combination with other air pollutant."

In setting standards that are "requisite" to protecting public health and welfare, as provided in Section 109(b), EPA's task is to establish standards that are neither more nor less stringent than necessary for these purposes. In so doing, EPA may not consider the costs of implementing the standards (*Whitman v. American Trucking Associations*, 531 U.S. 457, 465–472, 475–76 [2001]).

Section 109(d)(1) requires that "not later than December 31, 1980, and at 5-year intervals thereafter, the Administrator shall complete a thorough review of the criteria published under Section 108 and the national ambient air quality standards . . . and shall make such revisions in such criteria and standards and promulgate such new standards as may be appropriate. . . ." Section 109(d)(2) requires that an independent scientific review committee "shall complete a review of the criteria . . . and the national primary and secondary ambient air quality standards . . . and shall recommend to the Administrator any new . . . standards and revisions of existing criteria and standards as may be appropriate. . . ." Since the early 1980s, this independent review function has been performed by CASAC of EPA's Science Advisory Board (SAB).

2.2 Background on Previous NO_2 and SO_2 Secondary NAAQS Reviews

The current secondary NAAQS review for NO_x/SO_x will examine a number of issues that were of central importance in previous NAAQS nitrogen or sulfur reviews. For instance, in the previous review of the NO_2 NAAQS, completed in 1995, welfare effects were assessed primarily with respect to effects of ambient concentrations of NO_2 on vegetation by way of a literature review. A full risk assessment was not conducted for other welfare effects associated with NO_2. The decision was made to retain the current standard for NO_2 as the uncertainty and variability associated with the science at that time limited any other action. The 1995 NO_2 review did state, however, that "growing evidence does indicate that the impact of nitrogen deposition on sensitive aquatic ecosystems may be significant," that certain areas of the country, including "the Catskills, Northern Appalachians, Valley and Ridge Province, and Southern Appalachians all show some potential for chronic acidification due to NO_3^-" (U.S. EPA, 1995). Additional topics highlighted within the 1995 NO_2 review also included the importance of acid neutralizing capacity (ANC) in surface water acidification, the influence of atmospheric nitrogen to eutrophication of the Chesapeake Bay, and the growing body of research on developing "critical loads" and "target loads" for nitrogen in various ecosystems.

In the previous review of the SO_2 NAAQS, completed in 1996, vegetation damage (growth, yield, and foliar injury) due to short-term and long-term exposures to SO_2 were avoided by maintaining the current secondary 3-hour standard (0.053 ppm) (U.S. EPA, 1982). Data on the effects of long-term exposures affecting species richness and species diversity, reduced growth, and premature needle drop were considered "weak and not developed well enough to provide the principal basis for selecting the level of a long-term SO_2 standard" (U.S. EPA, 1982). The 1982 staff paper did note, however, that long-term SO_2 concentrations may be affecting lichen and mosses, and that these should be considered in the larger context of regional acid deposition. (Note: the 1986 Addendum (U.S. EPA, 1986) and 1994 Supplement (U.S. EPA, 1994) solely addressed human health effects.)

The planned risk/exposure assessment described in this Scope and Methods Plan builds upon the methodology and lessons learned from the previous NAAQS reviews. This plan is based on our current understanding of the NO_x and SO_x scientific literature and is subject to change as the NO_x/SO_x ISA undergoes revision. Currently, the EPA's Office of Research and Development's (ORD) National Center for Environmental Assessment (NCEA) has compiled and synthesized the most policy-relevant science available to produce a draft of the ISA, which has been used in the development of the approach described here. The approach described in this plan may also be modified according to CASAC and public comments following their review of this document as well as any additional information contained in the final version of the ISA.

2.3 Overview of Ecological Risk Assessment and the Scope of the Secondary NAAQS Review for NO_x and SO_x

The conventional framework for ecological risk assessment consists of three phases:

- Problem Formulation,
- Analysis, and
- Risk Characterization.

These phases have been described in more detail in the *Guidelines for Ecological Risk Assessment* (U.S. EPA, 1998) and other documents prepared by the Risk Assessment Forum (U.S. EPA, 1991, 1992a, 1992b). Generally, the Problem Formulation Phase describes the goals, breadth, and focus of the assessment including assessment endpoints, data needs, and anticipated analyses. It results in three products: (1) assessment endpoints that adequately reflect management goals of the ecosystem they represent, (2) conceptual models that describe key relationships between stressor and assessment endpoint or between several stressors and assessment endpoints, and (3) an analysis plan. The Scope and Methods Plan for the secondary

NO_2/SO_2 NAAQS Review represents the Problem Formulation Phase of the ecological risk assessment framework. The Analysis Phase of an ecological risk assessment might include environmental exposure profiles, the magnitude of spatial and temporal patterns of exposure, and summaries of the data analyses on effects and their association with assessment endpoints. The Risk Characterization Phase of an ecological risk assessment may be a quantitative or qualitative assessment that integrates exposure and effects profiles and estimates risks by categories, such as individuals or populations via modeling techniques. In the risk/exposure assessment, EPA plans to draw upon the ISA to develop quantitative and qualitative estimates of the risks of adverse welfare effects occurring as a result of current ambient levels of nitrogen oxides and sulfur oxides, levels that meet the current standards for NO_2 and SO_2, or levels that meet possible alternative standards. The issues that are addressed by the Analysis Phase and Risk Characterization Phase are part of the Risk/Exposure Assessment for the Secondary NAAQS Review for NO_x and SO_x.

Welfare impacts from air pollution to ecosystems are a serious concern. There are many harmful environmental effects of air pollution, including acid rain, ozone formation, decreased visibility, and effects on climate. In addition to direct adverse impacts to the biological and biogeochemical components of natural ecosystems, these direct ecological impacts can affect the welfare amenities of ecosystems in terms of their aesthetic and recreational amenities. Impacts on climate are also considered welfare effects, such as those due to nitrous oxide, N_2O. Welfare effects from the production of greenhouse gases such as N_2O can also cause significant impairment of ecosystems through climate change processes.

Against this broad background to welfare impact issues, this risk/exposure assessment will focus on ecological quality and its effects from acidification and enrichment related to nitrogen and sulfur air pollutants. In previous secondary reviews, acidification was evaluated for its damage to materials, including decay of buildings, statues, and sculptures that are part of our national heritage. The current assessment will focus on the influence of acid deposition on soil, forests, and waterbodies. Nitrogen deposition may also contribute to eutrophication (oxygen depletion) of water bodies, the symptoms of which include algal blooms (some of which may be toxic), fish kills, and loss of plant and animal diversity. These ecological changes impact human populations by changing the availability of seafood and creating a risk of consuming fish or shellfish contaminated from toxins produced by algal species that may be at an advantage in eutrophic systems as mentioned in the ISA. This reduces our ability to use and enjoy our coastal ecosystems, and causes an economic impact on people who rely on healthy coastal ecosystems, such as fishermen and those who cater to tourists.

Visibility impairment and ozone formation are additional welfare effects that are being addressed in the particulate matter (PM) and ozone NAAQS reviews. Visibility is being addressed in a separate secondary review effort as part of the PM NAAQS standard review. Ozone pollution's secondary impacts include damage to plant vegetation. Ozone is a secondary product of precursors NO_2 and volatile organic carbon. A separate rule review effort is in place for ambient ozone concentration, and its secondary impacts will also not be addressed in this NO_x and SO_x risk/exposure assessment.

In this current review, the appropriateness of NO_2 as an indicator for NO_x species and SO_2 as an indicator for SO_x species will be evaluated. This review will evaluate new information published in the peer-reviewed literature since the completion of the last NO_2 (1995) and SO_2 (1996) reviews, including assessments of the adequacy of the current secondary NAAQS, consideration of whether there is a possible need for a new single indicator or suite of indicators, as well as changed or retained level(s) and/or averaging times for the standards, which may include nitrogen and sulfur compounds other than NO_2 and SO_2.

This Scope and Methods Plan is intended to facilitate consultation with the CASAC, as well as the public, and to obtain advice on the overall scope, approaches, and key issues in advance of the completion of such analyses and presentation of results in the first draft of the risk/exposure assessment. The risk/exposure assessment is intended to be a tool that, together with other information contained in the NO_x/SO_x ISA, can aid the Administrator in judging whether the current secondary standards are requisite to protect public welfare from any known or anticipated adverse effects, or whether these standards should be retained, revised, revoked and/or replaced with alternative standard(s) having different indicators to provide the required protection.

2.3.1 Overview of Risk Assessment Framework for Deposition-related Ecological Effects

The risk/exposure assessment framework is intended to serve as a conceptual map of the analytical and decision steps necessary to estimate the ecological risks associated with alternative forms and levels of standards for NO_x and SO_x. As noted in the Integrated Review Plan for this review (U.S. EPA, 2007a), the purpose of the risk/exposure assessment is to assess the potential adversity of impacts including the effects of the pollutants on ecosystem goods and services, the degree to which ecosystem functions are impaired, long-term trends in specific ecosystems (where available), and both monetized and non-monetized valuation of ecosystem services. The ability of the risk/exposure assessment to characterize these impacts will depend on a number of factors, including the state of the supporting science, the availability of data on ecosystem baseline conditions and/or responsiveness to changes in nitrogen and sulfur deposition, and the time available under the regulatory development process.

The design of a risk assessment framework is discussed in the 1998 *Guidelines for Ecological Risk Assessment* (U.S. EPA, 1998). For this NO_x/SO_x risk/exposure assessment, EPA designed a flow diagram that represents how nitrogen and sulfur compounds move from "source to dose" in the environment. (See **Figure 2-1**.) This diagram represents the risk assessment framework for deposition-related ecological risks. It consists of two general activities: 1) characterization of exposure and 2) characterization of effect. More specifically, this framework depicts the processes and transformations among atmospheric concentrations, deposition, ecosystem impacts, exposure to biologically relevant species, and ecosystem responses via ecosystem services and valuation.

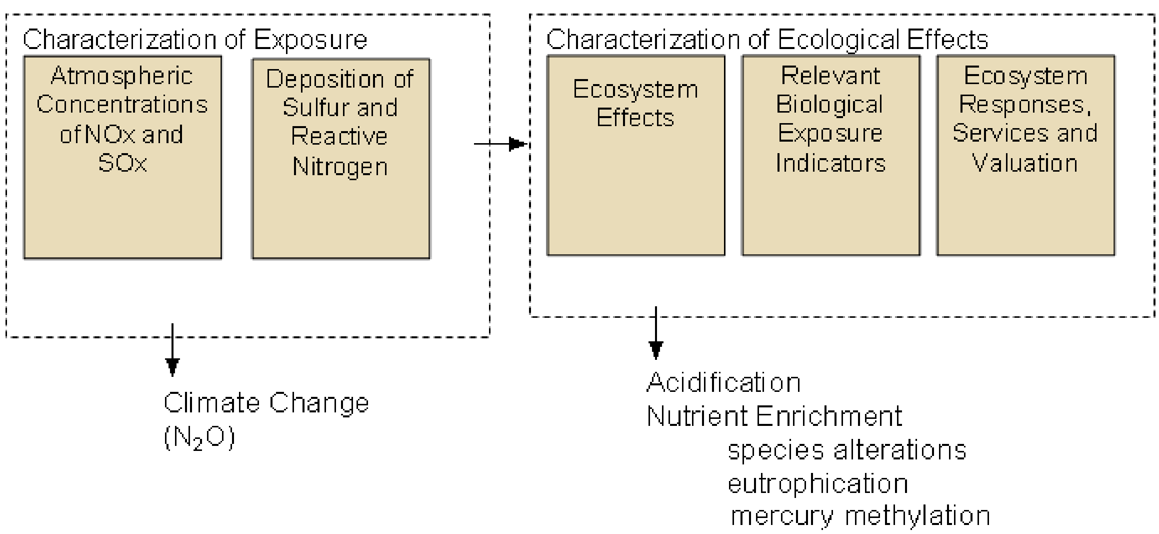


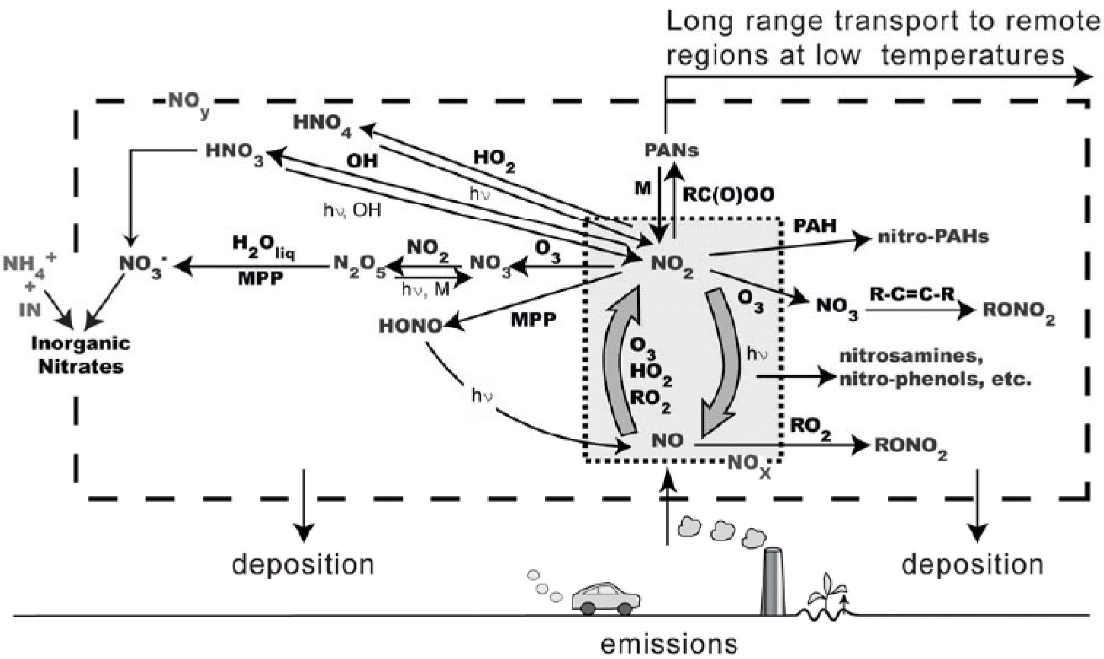
Figure 2-1. Risk assessment framework for deposition-related ecological risks.

The ecosystem response may be expressed as an "endpoint" in the ecological risk assessment. For this review, the level of acidification and nitrogen and/or sulfur enrichment that results in a harmful effect may be deemed an endpoint. This risk assessment framework entails modeling to calculate ecosystem loading and exposure required to reach the endpoint. This load is also known as the critical load. A critical load analysis is a form of site-specific risk assessment. It considers exposure to and response by various ecosystem receptors to identify the amount of atmospheric deposition (or loading) above which adverse ecological effects occur, serving as the endpoint of the risk assessment. Critical load analysis, within a risk assessment framework, can be a potential indicator for policy analysis.

Each component of the framework has a number of decisions required in implementing a framework to analyze alternative air quality standards. In an analytical world unconstrained by data and resource limitations, one could envision a nationwide comprehensive risk assessment covering all potentially affected sensitive ecosystems and all scientifically supported effects. However, as noted by the Science Advisory Board in their recent review of the analytical plan for the 2nd Prospective Analysis of the Costs and Benefits of the Clean Air Act, "a comprehensive quantitative national assessment of the ecological benefits of the CAA Amendments is not a realistic expectation." We recognize this limitation in developing the framework for the NO_x/SO_x risk/exposure assessment, and in the following sections, we describe a detailed framework for assessing ecological risks that follows the principle of obtaining the maximum amount of policy-relevant risk information possible given the data, resources, and time limitations.

2.3.2 Overview of Nitrogen Deposition

The sum of mono-nitrogen oxides, NO_2 and NO, typically are referred to as nitrogen oxides (NO_x) in the atmospheric science community. More formally, the family of nitrogen oxides includes any gaseous combination of nitrogen and oxygen, e.g., NO_2, NO, N_2O, N_2O_3, N_2O_4, and N_2O_5. Total reduced nitrogen (NO_y) includes all nitrogen oxides as well as gaseous and particulate nitrate species such as HNO_3, PAN, and aerosol phase ammonium nitrates. Reduced atmospheric nitrogen species include ammonia gas (NH_3) and ammonium ion (NH_4^+), the sum of which is referred to as NH_x. Atmospheric nitrogen deposition often is delineated farther as dry (gas and particulate phases) or as wet (precipitation derived ion phase). (See **Figure 2-2**.)

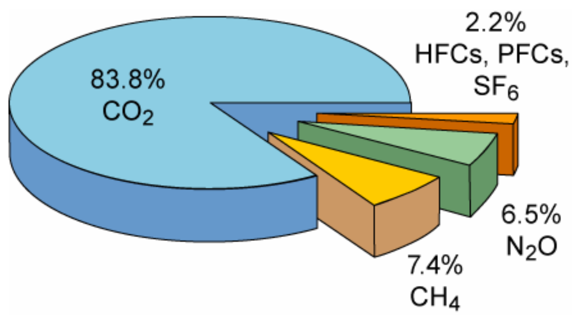


(Source: U.S. EPA, 2007e. ISA for Oxides of Nitrogen and Sulfur Environmental Criteria. December 2007, EPA/600/R-07/145A).

Figure 2-2. Schematic diagram of the cycle of reactive, oxidized nitrogen species in the atmosphere. (IN refers to inorganic particulate species (e.g., Na+,Ca++), MPP to multiphase processes, hv to a solar photon, and R to an organic radical. Particulate phase organic nitrates are also formed from the species on the right side of the figure.)

N_2O has not been considered in setting previous NO_2 NAAQS standards. In the first NO_x review, N_2O was not considered an air contaminant because there was "no evidence to suggest N_2O is involved in photochemical reactions in the lower atmosphere" (U.S. EPA, 1971). N_2O was addressed in both the 1982 and 1993 criteria documents. In 1982, it was described as one of the eight nitrogen oxides that may be present in the ambient air, but "not generally considered a pollutant." The effect of N_2O on stratospheric ozone was described, and the criteria document noted that N_2O may cause a small decrease in stratospheric ozone (U.S. EPA, 1982). Finally, the criteria document also concluded that N_2O significantly contributes to the atmospheric greenhouse effect by trapping outgoing terrestrial radiation, and that the issue was being investigated, but that many years of research were still needed to reliably assess the issue. In 1993, the criteria document again identifies N_2O as an oxidized nitrogen compound that is not generally considered to be an air pollutant, but does have an impact on stratospheric ozone and is considered among the more significant greenhouse gases. These documents clearly consider N_2O as within the scope of the listed nitrogen oxides criteria pollutant.

The 2007 draft of the ISA (see Sections 2.2, 3.1 and 4.4 in U.S. EPA, 2007b) acknowledges N_2O as a potent greenhouse gas and discusses N_2O sources and emissions in the United States, as well as the biogeochemistry of the microbial mediated production via denitrification in natural ecosystems. Based on the current U.S. Greenhouse Gas (GHG) Inventory (U.S. EPA, 2007d), nitrous oxide contributes approximately 6.5 % to total greenhouse gas emissions (in CO_2 equivalents) (**Figure 2-3**).



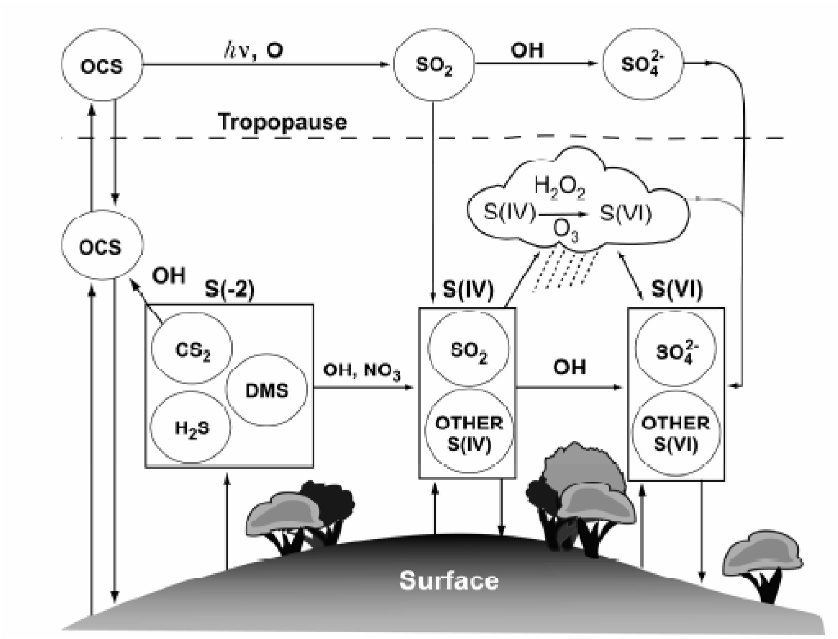
Source: U.S. EPA, 2007d

Figure 2-3. Percent of Total U.S. Emissions of greenhouse gases in CO_2 equivalents.

Since the definition of “welfare effects” includes effects on climate [CAA Section 302(h)], we will include N_2O within the scope of this review. However, it is most appropriate to analyze the role of N_2O in anthropogenic climate change in the context of all of the greenhouse gases. Since that is outside the scope of this review, it will not be a quantitative part of this assessment.

2.3.3 Overview of Sulfur Oxides and Sulfur Deposition

SO_2 is one of a group of substances known as SO_x, which include multiple gaseous (e.g., SO_2, SO, SO_3, S_2O_3, S_2O_7) and particulate (e.g., ammonium sulfate) species (**Figure 2-4**). Acidification can result from the atmospheric deposition of SO_x and NO_x; in acid deposition, these species combine with water in the atmosphere to form sulfuric acid (H_2SO_4) and HNO_3. Acidification is an environmental effect in which acid precipitation lowers the natural pH of waterbodies and/or damages terrestrial ecosystems. Over the past few decades, acidification of waterbodies has been recognized as an environmental issue throughout Europe and North America, and steps have been taken to control SO_x and NO_x emissions and to identify the recovery of the impacted ecosystems. Due to known acute effects on plants, in previous NAAQS reviews, SO_2 served as the chemical indicator for SO_x species.



(Source: U.S. EPA, 2007e. ISA for Oxides of Nitrogen and Sulfur Environmental Criteria. December 2007, EPA/600/R-07/145A [adapted from Berresheim et al., 1995]).

Figure 2-4. Transformation of sulfur compounds in the atmosphere.

2.3.4 Targeted Effects for This Risk/Exposure Assessment

The two classifications of effects that are targeted for this risk/exposure assessment are acidification and nitrogen and sulfur enrichment. Both effects occur in response to deposition of NO_x and SO_x. Acidification effects can occur from either nitrogen or sulfur deposition, and the relative contribution of each type of deposition depends on the characteristics of the affected ecosystem (see Section 4.2 of the ISA, U.S. EPA, 2007b). Nitrogen and sulfur enrichment represents a continuum of effects, and it can be characterized as a positive or negative effect, depending on the selected endpoint, location and baseline conditions of an ecosystem. Enrichment effects are caused by nitrogen or sulfur deposition, but are dominated by nitrogen deposition, which will be the focus of the risk/exposure assessment. Nitrogen enrichment in ecosystems may alter the native terrestrial species composition (i.e., from wildflower meadows to shrubs), and can result in eutrophication in aquatic systems (see Section 4.3 of the ISA, U.S.

EPA, 2007b). Thus, the framework for this review highlights four main areas that will be evaluated for a risk/exposure assessment in this plan:

1. Risks to terrestrial ecosystems from nitrogen enrichment effects
2. Risks to aquatic ecosystems from nitrogen enrichment effects (eutrophication)
3. Risks to terrestrial ecosystems from acidification effects (nitrogen and sulfur)
4. Risks to aquatic ecosystems from acidification effects (nitrogen and sulfur)

In addition to the four targeted effects listed above, we will address, as appropriate, impacts associated with nitrous oxide and the influence of sulfur enrichment on methylmercury production. **Figure 2-5** provides a conceptual diagram of the processes we will need to model in our risk and exposure assessment. Atmospheric fate and transport is the initial point of departure for the analysis, and is fully integrated in the treatment of NO_x, other reactive nitrogen species, and SO_x. The results of the atmospheric fate and transport process are atmospheric loading of sulfur and total reactive nitrogen, as well as concentrations of N_2O. As mentioned earlier, a quantitative assessment of N_2O is not currently within the scope of this review.

Atmospheric loadings of total reactive nitrogen and SO_x lead to deposition of nitrogen and sulfur to terrestrial and aquatic ecosystems. In addition to the major ecological effects of acidification and nutrient enrichment, sulfur deposition also leads to enhanced methylmercury (MeHg) production in aquatic systems and, in turn, increases the risks for bioaccumulation and biomagnifications of mercury in food chains (see Section 4.4 in U.S. EPA, 2007b). This interaction between sulfur deposition and methylmercury production can exacerbate an already important mercury problem, especially in coastal and eutrophic waters subject to hypoxic algal blooms. The focus of the quantitative risk/exposure assessment will be the set of effects associated directly with acidification and nitrogen and sulfur enrichment, but we will also provide a qualitative assessment of potential impacts of sulfur deposition on waterbodies that are vulnerable to increased mercury methylation.

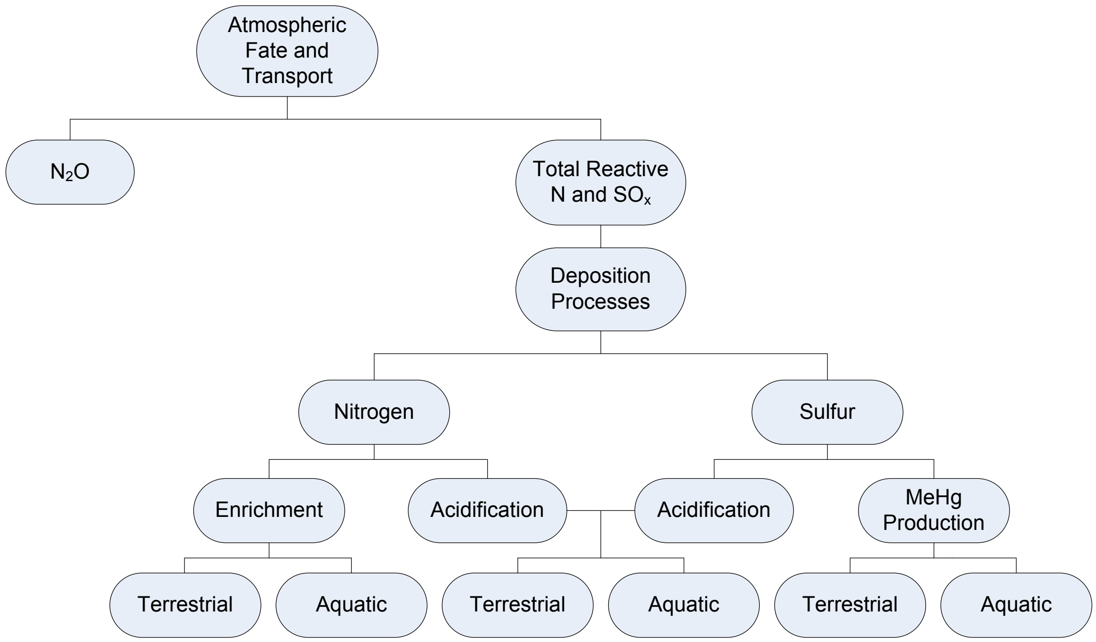


Figure 2-5. General flow of processes addressed in the risk/exposure assessment for NO_x and SO_x secondary standards.

2.4 Key Policy Relevant Questions

The 2007 Integrated Review Plan of the Secondary NAAQS for NO_2 and SO_2 introduced a series of policy-relevant questions to frame the approach EPA will take in this review (U.S. EPA, 2007a). The Review Plan indicated that issues of ecosystem susceptibility should be addressed, as well as the issue of whether individual effects or combined effects are more important to a given ecosystem (i.e., is it NO_x or SO_x acting individually that is important, or is it the combination of NO_x and SO_x that needs to be addressed). For example, both NO_x and SO_x are associated with acidification effects, while nitrogen is associated with nutrient enrichment and eutrophication effects, and sulfur is associated with increased mercury (Hg) methylation.

Both EPA and CASAC have acknowledged the importance of NO_x, SO_x, and their associated transformation products with respect to acidification effects on ecosystems. This review will focus on the ecosystem-related welfare effects that result from the deposition of these pollutants and their transformation products, rather than on the effects of aerosol NO_x and SO_x that remain in the atmosphere.

For this secondary NAAQS review of NO_x/SO_x, the primary policy-relevant questions include:

- What are the known or anticipated welfare effects influenced by ambient NO_x and SO_x, and for which effects is there sufficient information available to be useful as a basis for considering distinct secondary standard(s)?
- What is the nature and magnitude of ecosystem responses to NO_x and SO_x that are understood to have known or anticipated adverse effects, and what is the variability associated with those responses (including ecosystem type, climatic conditions, environmental effects, and interactions with other environmental factors and pollutants)?
- What are the biologically relevant indices that adequately capture the relationship between ecosystem exposure and response for the known or anticipated welfare effects we are trying to protect?
- To what extent do receptor surfaces influence the deposition of gases and particles (dry deposition), since dry deposition can contribute significantly to total deposition?
- What are the appropriate air quality indicator(s), averaging time(s), form(s), and level(s) of standards that are requisite to avoid those ecosystem responses?
- To what extent do the current standards provide the requisite protection for the public welfare effects associated with NO_x and SO_x?
  - Should the current secondary standards for NO_2 (as an indicator of NO_x) and SO_2 (as an indicator for SO_x) be retained, revised, or revoked and/or replaced with alternative standard(s) having different indicators to provide the required protection from known or anticipated adverse public welfare effects?
  - Can effects from NO_x be distinguished from effects due to total reactive nitrogen?

To the extent that the evidence suggests revision of the current secondary NO_x/SO_x NAAQS is appropriate, ranges of standards will be identified (including different or alternate indicators, terms of exposure indices, averaging times, levels, and forms) that reflect a range of alternative policy judgments as to the degree of protection that is requisite to protect public welfare from known or anticipated adverse effects. To account for variability in ecosystem responses and land uses across the nation, ecosystem characteristics may be an important consideration in evaluating the form(s) of the standards. The form(s) of the standard(s) may be based on a complex formula that incorporates ecosystem characteristics, land uses, atmospheric transformations, climatic conditions, environmental effects and other interactions. In so doing, the following questions should be addressed:

- Does the available information provide support for considering different NO_x/SO_x chemical indicators or exposure indices?
- Does the available information provide support for considering some joint standard(s) or are separate standards appropriate?
- What range of levels and forms of alternative standards are supported by the information, and what are the uncertainties and limitations in that information?
- To what extent do specific levels and forms of alternative standards reduce adverse impacts attributable to NO_x/SO_x, and what are the uncertainties in the estimated reductions?

In order to be able to answer these questions, we believe that the relevant scientific and policy issues that need to be addressed in the science, risk/exposure, and policy assessment portions of this review include:

- Identifying important chemical species in the atmosphere
- Identifying the atmospheric pathways that govern chemical transformation, transport, and deposition of NO_x and SO_x to the environment
- Identifying the attributes of ecosystem receptors that govern their susceptibility to effects from deposition of nitrogen and sulfur compounds
- Identifying the relationships between ambient indicators and biologically relevant indices of effects, including ecosystem services associated with the indicator (but not excluding other non-economic evaluations)
- Evaluating alternative measures to assess the adversity of effects on ecosystem services, including, for example, economic valuation
- Evaluating if current levels may have a long-term impact due to cumulative loadings, and if this is relevant to a NAAQS review
- Evaluating environmental impacts and sensitivities to varying meteorological scenarios and climate conditions

2.5 Proposed Schedule

The proposed schedule for the joint NO_2/SO_2 secondary NAAQS review is shown in **Table 2-1**; underlined dates indicate the court-ordered schedule. Consultation with CASAC and the public on the first draft of the ISA and this Scope and Methods Plan is planned for April 2008. Based on this consultation, the plan for the risk/exposure assessment may be revised as needed. The first draft of the risk/exposure assessment and the second draft of the ISA will be released to CASAC and the public in August 2008. EPA will receive comments on these draft documents from CASAC and the public at a meeting in October 2008. A revised risk/exposure assessment will be released in March 2009 followed by a CASAC and public review in May 2009. The final risk/exposure assessment for the secondary NO_2/SO_2 NAAQS review will be released in July 2009.

Table 2-1. Proposed Schedule for the Joint NO_2 and SO_2 Secondary NAAQS Review*

Stage of Review	Major Milestone	Draft Target Dates
Planning	Literature search	Ongoing
	Federal Register call for information	December 2005
	Prepare the draft NO_2/SO_2 NAAQS Work Plan	December 2005–August 2007
	Workshop on science/policy issues	July 2007
	CASAC consultation	October 2007
	Prepare the final integrated NO_2/SO_2 NAAQS Work Plan	December 2007
Integrated Science Assessment (ISA)	Prepare first draft of the ISA	December 2007
	CASAC/public review of the first draft of the ISA	April 2008
	Prepare the second draft of the ISA	July/August 2008
	CASAC/public review of the second draft of the ISA	October 2008
	Prepare the final ISA	<u>December 12, 2008</u>
Risk/Exposure Assessment (REA)	REA methodology released to the CASAC and the public	February 2008
	CASAC/public consultation on the REA methodology	April 2008
	First draft of the REA released to the CASAC and the public	August 2008
	CASAC/public review of the first draft of the REA	October 2008
	Second draft of the REA released to the CASAC and the public	March 2009
	CASAC/public review of the second draft of the REA	May 2009
	Final REA released	July 2009

Stage of Review	Major Milestone	Draft Target Dates
Policy Assessment/ Rulemaking	Publish ANPR	August 2009
	CASAC review/public comment on ANPR	October 2009
	Proposed rulemaking	February 12, 2010
	Final rulemaking	October 19, 2010

* Schedule may be modified, as necessary, to reflect actual project requirements and progress. Underlined dates indicate the court-ordered schedule.

3. ANALYSIS PLAN

Because ecosystems are diverse in biota, climate, geochemistry, and hydrology, response to pollutant exposures can vary significantly. Also, these diverse ecosystems are often neither abundant nor distributed evenly across the United States. To target acidification and nitrogen and sulfur enrichment, this Scope and Methods Plan focuses on four main ecosystem effects on terrestrial and aquatic systems identified in the ISA:

- Terrestrial nitrogen enrichment
- Aquatic nitrogen enrichment, including eutrophication
- Terrestrial acidification due to nitrogen and sulfur
- Aquatic acidification due to nitrogen and sulfur

In addition to these four effects, we plan to address, as appropriate and within our time constraints, impacts associated with nitrous oxide (N_2O)and the influence of sulfur enrichment on methylmercury production. Since these ecosystem effects are not found evenly distributed across the United States, we plan to perform risk/exposure assessment case studies for specific areas of the U.S. We plan to select these case studies from those areas of the United States where ecosystems are identified as sensitive to nitrogen and/or sulfur deposition effects.

Once those sensitive areas are determined, we will decide how best to design and conduct case study analyses that contain the sensitive ecosystems of interest. These locations may vary in size from a single site to a region containing numerous lakes. Methods of assessments can include cluster analyses of regions with common ecosystem sensitivities (e.g., lakes and streams of the Adirondack Mountains), site-specific quantitative modeling analyses, qualitative analyses, and review and summary of previous risk/exposure assessments. From these qualitative-quantitative analyses of sensitive areas and case studies in different regions of the U.S., we will discern if the results can be used for a broader characterization of national conditions to represent key components of our nation’s ecology. To be clear, this exercise is not a national-scale ecological risk assessment but, rather, is intended to be a qualitative analysis of multiple ecosystems’ quantitative-qualitative risk/exposure assessments.

The risk/exposure assessment for the Secondary NAAQS for NO_x and SO_x will build upon the scientific information presented in the 2007 draft ISA (U.S. EPA, 2007b). The ISA documents ecological effects of nitrogen and sulfur deposition, biogeochemical indicators of effects, and areas of the U.S. where ecosystem effects have been studied. The ISA also recommends selected indicators and case studies as candidates for risk/exposure assessment and ecosystem services valuation. In this section, we will identify and describe how those recommendations can be considered in the assessment in order to provide information for policy decision making.

The risk/exposure assessment will focus on ecosystem welfare effects that result from the deposition of total reactive nitrogen and sulfur. The anticipated spatial extent and diversity of ecological effects due to deposition of nitrogen and sulfur do not facilitate a nationwide analysis. Further, some areas of the United States are more vulnerable to the effects of deposition than others. Because of this diversity, we consider it valuable to formulate a strategy that is designed to protect sensitive systems, while allowing for flexibility in areas that are more resilient. As a result, this assessment intends to evaluate potential alternatives to current indices in an attempt to quantify the relationship between ambient concentrations of NO_x and SO_x and potential welfare effects. To create these indices, we plan to evaluate exposures and impacts in various ecosystem case studies with differing responses related to nitrogen and sulfur inputs and explore relationships between ambient concentrations and deposition of nitrogen and sulfur.

As previously described, deposition of SO_x and NO_x compounds affects ecosystems in various adverse ways and at different spatial and temporal scales in diverse regions of the country. In its review of the analytical plan for the 2nd Prospective Analysis of the Costs and Benefits of the Clean Air Act, the Science Advisory Board recommended that EPA consider including studies of upland as well as coastal sites, because air deposition is often not the primary contributor of nitrogen in coastal sites, while it may be the dominant source in upland locations. The Analysis Plan Phase of a risk assessment includes selecting data that will be used, analyzing exposure (including spatio-temporal conditions), analyzing effects, and summarizing conclusions about exposure.

In order to address the policy-relevant questions that are guiding the scope of this review, this risk/exposure assessment intends to evaluate the relationships between atmospheric concentrations, deposition, biologically relevant exposures, ecosystem effects, and ecosystem services.

To evaluate the nature and magnitude of ecosystem responses associated with adverse effects, the risk/exposure assessment plans to examine various ways to quantify the relationship between air quality indicators, deposition of biologically accessible forms of nitrogen and sulfur, biologically-relevant indices relating to deposition, exposure and effects on sensitive receptors, and related impacts to ecosystem change and services. To the extent feasible, the risk/exposure assessment should also evaluate the overall load to the system for nitrogen and sulfur as well as the variability in ecosystem responses to these pollutants. The assessment intends to determine the exposure metrics that incorporate the temporal considerations (i.e., biologically relevant timeframes), pathways, and biologically relevant indices necessary to maintain the functioning of these ecosystems. In addition, the risk/exposure assessment plans to evaluate the contributions of atmospherically deposited nitrogen and sulfur relative to total loadings in the environment. For the atmospheric contribution to total nitrogen, we also plan to evaluate the contribution of NO_x to total reactive nitrogen in the atmosphere relative to the contributions of reduced forms of nitrogen (e.g., ammonia, ammonium).

The scope of the risk/exposure assessment will depend, in part, on the answers to the following questions:

- What are the appropriate geographic scales and/or time frames for the risk assessment? Information that will be considered in addressing this question includes mapping datasets, research studies of sensitive ecosystems ranging in size from single lakes to stream and lake systems within a large geographic region (e.g., the Southern Appalachian Mountains), identification of representative ecosystem types, air pollution gradient studies, a weight-of-evidence approach incorporating both qualitative and quantitative information on risk, or some combination of the above.
- How can regional variation of effects be taken into account? How should the risk/exposure assessment address acidification and nutrient-enrichment effects in different areas and ecosystem types (e.g., mesic, arid, mountain forests, alpine, subalpine)?
- To what degree are assumptions supported by the available science regarding linkages between pollutants in ambient air, deposition, and measurable ecosystem effects,

including effects on ecosystem services? What are the most useful metrics of both ambient pollution and the resulting effects?

- To what degree should the risk/exposure assessment take the potential for recovery into account in selecting data for qualitative and quantitative assessments?
- How can uncertainties be minimized and appropriately characterized?

Because this risk/exposure assessment intends to focus on two basic secondary effects related to sulfur or nitrogen pollutants—acidification and enrichment—and because ecosystems may respond differently to these effects, it will be necessary to first perform risk/exposure assessment case studies unique to the effect and ecosystem type. We will assess the feasibility to consolidate effects and/or ecosystems in the risk/exposure assessment and, where feasible, perform a broader characterization. However, some ecosystems and their effects may be too unique to consolidate into a broad characterization.

Upon completion of all risk/exposure assessment case studies, the results of the assessments performed for unique combinations of effects and ecosystem types will be presented together to facilitate decision making on the total effects of nitrogen and sulfur deposition. Ecosystem services that relate to the effects will be identified and valued if possible. Ecosystem services provide an additional way to compare effects across various ecosystems. The following is an overview of the risk/exposure assessment process, as well as more in-depth discussions on topics addressed in the seven steps (see **Figure 3-1**).

Steps for Characterizing Ecological Effects

The seven basic steps guiding the plan for the overall the risk/exposure assessment and the assessments for each case study area of interest are shown in **Figure 3-1**. These seven steps capture the components of the risk assessment framework by addressing the selection of effects, indicators and ecosystem services measured for exposure via atmospheric deposition of total reactive nitrogen and sulfur from ambient air. The initial steps of identifying effects, sensitive ecosystems, and potential indicators have been performed and documented in the ISA. In addition, the ISA identifies and reviews candidate multimedia models available for fate and transport analyses of a variety of ecosystems. The science documented in the ISA will play a key role in planning and conducting the risk assessment. It is possible that, for some of the desired case study areas, data may not be abundant enough to perform a quantitative assessment for each of the steps; in those cases, we may choose to execute some of these steps in a qualitative or semi-quantitative fashion.

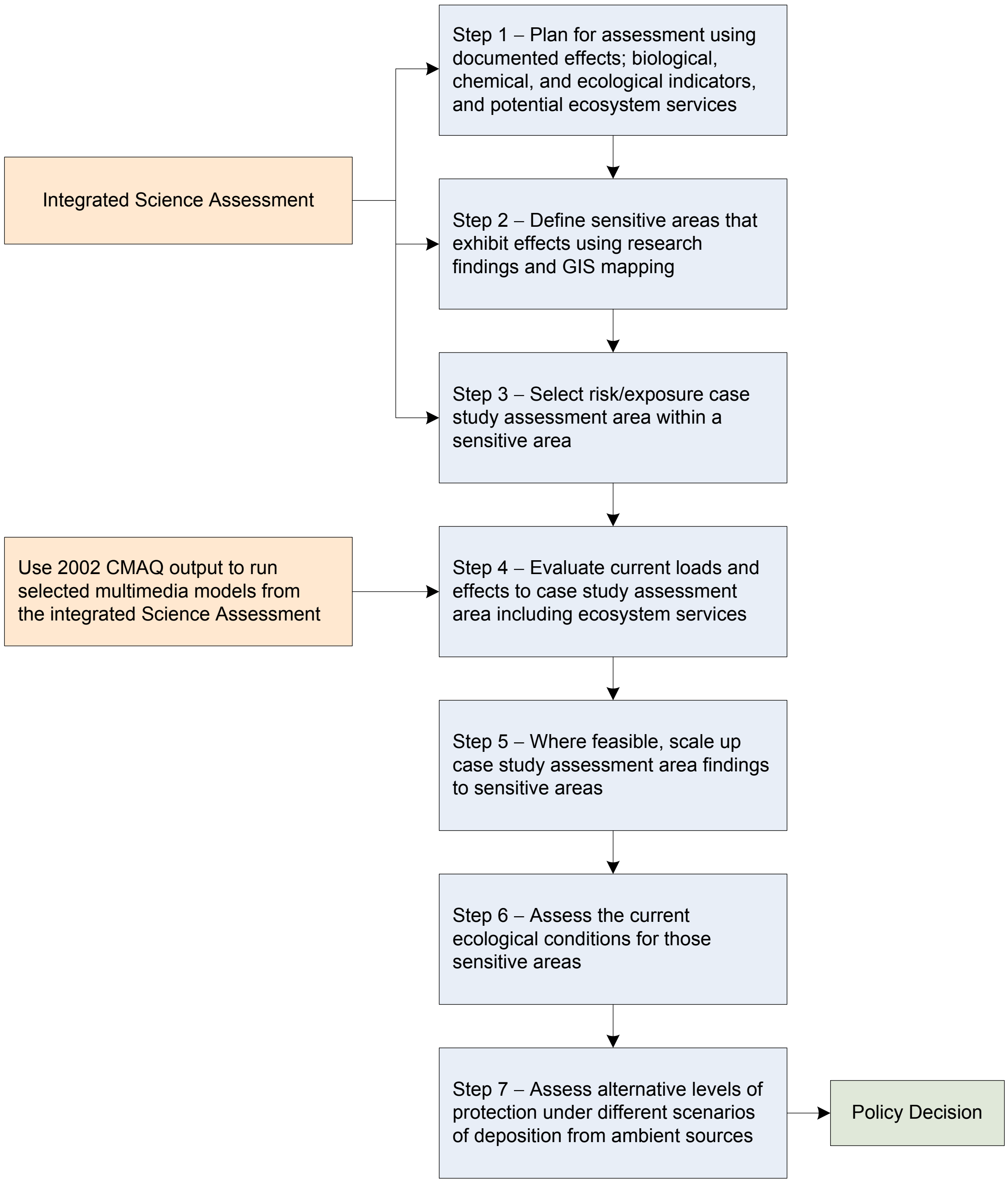


Figure 3-1. Seven step approach to planning and implementing risk/exposure assessment.

3.1 Step 1 – Plan for Assessment Using Documented Effects; Biological, Chemical, and Ecological Indicators, and Potential Ecosystem Services

3.1.1 Documented Effects and Relevant Indicators of Effects for Each Ecosystem, Including Function and Service

To assess the impacts of total reactive nitrogen and sulfur loading, we plan to identify adverse terrestrial and aquatic effects due nitrogen and sulfur deposition. The impacts of acidification, nitrogen-induced eutrophication, and changes in species diversity due to nitrogen and/or sulfur saturation are documented in EPA's 2007 ISA (U.S. EPA, 2007b). We plan to use this information to identify those ecosystems and ecosystem services that are considered most sensitive to acidification and nitrogen and sulfur enrichment and most informative for potential case study analyses in our overall risk/exposure assessment.

Environmental indicators are measures that track environmental conditions over time. EPA's Report on the Environment (ROE) (U.S. EPA, 2007c) defines an indicator as follows:

> A numerical value derived from actual measurements of a pressure, state or ambient condition, exposure, or human health or ecological condition over a specified geographic domain, whose trends over time represent or draw attention to underlying trends in the condition of the environment.

Indicators of ecosystem response include chemical, biological, and habitat measurements, such as forest extent and type, land cover, lake and stream acidity, nitrogen and phosphorus in streams, and contaminates in fish tissue.

Step 1 entails defining the biological and biogeochemical relevance of indicators for acidification and nitrogen and sulfur enrichment in order to select the most appropriate indicators for the risk/exposure assessment. The issue of which soil chemical and physical characteristics are most appropriate and the spatial variability in these characteristics will be addressed. For sulfur, the adsorption/desorption responses in soils may be important. In addition, we plan to consider the contribution of any internal sulfur sources (both organic sulfur mineralization and inorganic sulfur mineral weathering) to sulfate fluxes in soil, and the resultant differences in responses to decreases in sulfur deposition in surface and ground waters. For nitrogen, more attention will be focused on what soil features should be tracked; including how organic matter affects microbial processes. This characterization will also be linked to the role of vegetation, not only with respect to nitrogen cycling, but also in affecting organic matter quality via organic matter inputs to the soil. More detail on this approach to identifying indicators and a preliminary list of recommended indicators is provided below.

Preliminary NO_x and SO_x Indicators: In the ISA, relevant indicators were divided into one or both of the response categories: acidification or nutrient enrichment. **Table 3-1** presents acidification or nutrient enrichment indicators.

Table 3-1. Summary of Indicators Categorized by Effect

Acidification	Ecosystem Type	Nutrient Enrichment	Ecosystem Type
Acid stress index	A	Carbon budget (growth, carbon fixation, and respiration)	T
Acid neutralizing capacity	A, T	Concentration of chlorophyll	A, T
Alkalinity	A	Concentration of carotenoids	T
Aluminum, mobilization	A	Nitrogen, concentrations	A
Carbon to Nitrogen ratio	T	Photosynthetic rate	A, T
Calcium and magnesium concentrations	T	Phosphorus concentrations	T
Community structure	A, T	Species richness	A, T
Condition factor	A	Taxonomic density	A, T
Dissolved organic carbon	A	Thallus density	T
Dissolved organic nitrogen	A	Transpiration rate	T
Exchangeable cations	T		
Forest health	T		
Index of biotic integrity	A, T		
Metal mobilization	T		
pH	A, T		
Soil-base saturation	T		
Species composition	A, T		
Taxonomic richness	A, T		

A = Aquatic
T = Terrestrial

Key Acidification Indicators: **Table 3-2** presents key indicators that play a significant role in surface water acidification and recovery (summarized from Skjelkvale et al., 2005). Specific indicators of acidification are discussed in more detail below.

Table 3-2. Key Indicators of Acidification Due to NO_x and SO_x

Key Indicator Group	Examples of Indicators	Description
Acid anions	SO_4^{2-}, NO_3^-	Trends in these concentrations reflect recent trends in atmospheric deposition (especially SO_4^{2-}) and in ecosystem responses to long-term deposition (notably NO_3^- and desorbed SO_4^{2-}).
Base cations	Ca^{2+}, Mg^{2+}, $\Sigma(Ca^{2+}+Mg^{2+})$	These cations are mobilized by weathering reactions and cation exchange. These respond indirectly to decreases in SO_4^{2-} and NO_3^- because a reduced input of acids will lead to a reduction of neutralizing processes in the soil, thereby reducing the release of base cations to soil- and runoff waters.
Acidity	pH, (Gran) alkalinity, ANC	These indicators reflect the outcomes of interactions between changing concentration of acid anions and base cations.
Organic acidity	Dissolved organic carbon (DOC), total organic carbon (TOC)	Organic acids are common natural sources of acidity in surface waters.
Metals	Al, iron (Fe)	These metals are mobilized as a response to the deposition of SO_4^{2-} and NO_3^-.
Biological	Forest health, community structure, species composition, taxonomic richness, Index of Biotic integrity	Ecological effects occur at four levels: individual, population, community, and ecosystem. Metrics have been developed for each level to assess the adverse effects of acids.

ANC is an acidification indicator with relevance to soils, terrestrial ecosystems, and aquatic ecosystems and is a key indicator recommended in the ISA. ANC data are widely available for use in a risk/exposure assessment. Other indicators may be used in relation to particular ecosystems or specific sensitive areas. Chemical indicators such as pH or cation exchange capacity (CEC) are more widely available at the present time than the biological indicators.

Key Indicators of Nutrient Enrichment: Major indicators for nutrient enrichment to aquatic and terrestrial systems from air deposition of reactive nitrogen involve measurements based on available monitoring stations for wet deposition (NADP/national trend network [NTN]) and limited networks for dry deposition (CASTNET). Wet-deposition monitoring stations can provide more information on an extensive range of nitrogen species than is possible for dry-deposition monitoring stations. This creates complications in developing estimates for total nitrogen deposition levels because dry-deposition data sources will likely be underestimated. Individual studies measuring nitrogen deposition to terrestrial ecosystems that involve throughfall estimates for forested ecosystems can provide better approximations for total nitrogen deposition levels, but such estimates and related bioassessment data, are not available for the entire country. For terrestrial ecosystems, low calcium to nitrogen ratios in soils are commonly related to increased nitrification and potential increases in soil acidity and releases in NO_3 to receiving waters; however, these measurements are not always widely available.

For aquatic ecosystems, the indicators for "nutrient enrichment" effects reflect a combination of inputs from all media (e.g., air, discharges to water, diffuse runoff, groundwater inputs). Major aquatic system indicators involve nutrient loadings (Heinz Center, 2006), indicators of excess algal standing crops (U.S. EPA, 2006), or, in larger waterbodies, anoxia/hypoxia in bottom waters. (See **Table 3-3**). For nitrogen, loadings or concentration values related to total nitrogen (a combination of nitrates, nitrites, organic nitrogen, and total ammonia) are encouraged for inclusion in numeric criteria as part of EPA-approved state water quality standards (U.S. EPA, 2000). Given the nature of the major indicators for atmospheric deposition and indicators for aquatic and terrestrial ecological systems, a data-fusion approach that combines monitoring indicators with modeling inputs and outputs is often used (Howarth, 2007).

Table 3-3. Key Indicators of Nutrient Enrichment Due to NO_x and SO_x

Key Indicator Group	Examples of Indicators	Description
Nitrogen deposition	Nitrate or ammonia	From wet or dry deposition monitoring stations and networks.
Nitrogen throughfall deposition	Nitrate, ammonia, organic nitrogen	Special measurements in terrestrial ecosystem with corrections for nitrogen intercepted by plant canopies.
Nitrogen loadings and fluxes to receiving waters	Total nitrogen or constituent species combined with flow data from gauged stations	Reflects a combination of inputs from all media (air, discharges to water, diffuse runoff, and groundwater inputs). Relative role of air deposition should ideally be compared with air deposition data and also with available (preferably multi-media) models.
Other indicators of aquatic system nutrient enrichment (eutrophication)	Algal standing crop (plankton and periphyton); anoxia/hypoxia for estuaries and large rivers	Reflects a combination of inputs from all media (air, discharges to water, diffuse runoff, and groundwater inputs). Relative role of air deposition should ideally be compared with air deposition data and also with available (preferably multi-media) models.

3.1.2 Potential Ecosystem Services

We plan to identify the primary ecosystem service(s) for each of the effect classes (e.g., acidification or enrichment) and for major ecosystem types and components (e.g., terrestrial ecosystems, soils, aquatic ecosystems). These services may be characterized as: supporting services that are necessary for all other services (for example primary production), provisioning services (food, fuelwood), regulating services such as climate regulation or flood control, and cultural services including spiritual or religious values, aesthetic values, recreation values among others.

Define Change for Each Ecosystem Service: We intend to characterize the type of change, positive or negative, for each ecosystem service. This will be expressed in different ways, relative to the type of environmental system.

Identify the Indicator of Change: We plan to identify indicators for the major types of change. These indicators may use chemical/physical properties (e.g., ANC), or they may involve biological endpoints (e.g., bioassessment metrics, such as a fish or benthic invertebrate Index of Biotic Integrity [IBI]).

Identify Databases of Indicator Conditions: The indicators selected will relate to available compendiums of literature abstracts or actual database systems (as stand alone files or accessed through Web portals) to provide readily available and transparent ways to document the nature of the indicators and the indicator conditions used to define the environmental impairments.

Identify and Address Temporal Issues: Different ecoregions or biological provinces may understandably display differing degrees of susceptibility to impairments or differing recovery potential, depending on past land use or pollution histories. Some ecological systems may be capable of fairly rapid recovery responses once pollutant loadings are significantly abated; other systems, such as larger estuarine aquatic systems, may require much longer recovery times. These temporal issues will be documented in the analyses as possible.

3.2 Step 2 – Define Sensitive Areas That Exhibit Effects Using Research Findings and GIS Mapping

3.2.1 Defining Sensitive Ecosystems

Some ecosystems and areas of the United States are more sensitive than others to the effects of nitrogen and sulfur deposition (i.e., acidification and nitrogen and sulfur enrichment). In the risk/exposure assessment, we plan to begin with those ecosystems and case study areas identified in the ISA and consider potential near-field and far-field linkages.

Identify Biological, Biogeochemical, and Physiographic Linkages in These Ecosystems: Linked systems will be identified (e.g., upland terrestrial/aquatic areas linked to downstream estuarine system) where possible. Especially for larger watershed or basin-scale systems, some components of these study areas (e.g., estuaries linked to inland fluvial drainage areas) should show both direct near-field effects from nitrogen or sulfur enrichment as well as linked, far-field effects related to loadings from the inland drainage areas (**Figure 3-2**).

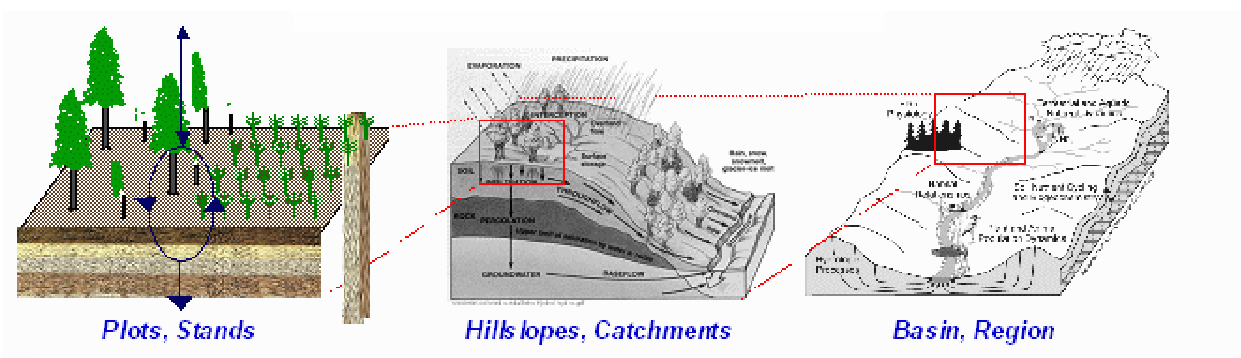


Figure 3-2. Ecosystem effects may range from near-field to far-field linkages.

Assess Significance of Linkages: The spatial extent of linkages supports different scales of risk assessment. For example, nutrient criteria could serve as the anchor of a broader-scale characterization of U.S. aquatic systems. Inland acid-sensitive waters in the eastern United States and nitrogen-sensitive ecosystems in the Rocky Mountains and other parts of the western United States may support a large-scale, special-area assessment. Ecosystem effects in special areas, such as the Adirondack Mountains or Class I areas of the United States may support area-level assessments. If the linkages are geographically significant, we plan to evaluate and determine the maximum scale of the case study area based on the linkages. The resultant case study area may be a common ecosystem, a subbasin, a riverbasin, or an airshed, and may be local, subregional, or regional in scale. If an area is identified as not having important linkages, then that area may be a good candidate for a case study area (e.g., local research of MeHg formation in Devil's Lake, WI).

Of special interest will be the characterization of linkages that reflect conditions for all parts of the country. For instance, national nutrient criteria for rivers and lakes can provide the foundation for national-scale characterizations involving nitrogen-enrichment effects for aquatic systems related to loadings from air deposition and other nitrogen sources. Available information on the nitrogen sensitivity of estuarine systems can further extend the scope of these analyses to include estuarine and other near-coastal waters. Information that deals with special case study areas (e.g., the Adirondack Mountains or special alpine and subalpine ecosystems in the West) that reflect impacts affecting sizeable regions may also be of interest. **Figure 3-3** provides examples of significant linkages.

3.2.2 GIS Mapping

To describe the national picture, we plan to map the locations of those sensitive ecosystems identified in Section 3.2.1 and identify the characteristics of the biological and biogeochemical properties that create the sensitivity. Identifying the key properties of sensitive systems may aid in estimating the sensitivity of currently unmapped areas. Sensitive areas can be identified at different spatial scales by using different approaches for defining the boundaries of the mapped units. Flexibility in the way sensitive areas are identified will enhance the utility of the highest-quality data. Such flexibility also facilitates identifying important ecological services or related welfare values of these sensitive natural systems. For instance, primary data collections are often made at the spatial level of small plots or stands. Geographic Information System (GIS)–based spatial analysis tools may be used to gain more insights from the collection of data. Models are often used to supplement direct measurements for larger spatial units, which may be reasonably localized catchments or land-cover patches. Integrated systems using monitoring, modeling, and interpolation approaches are often applied for larger watershed units or physiographical areas related to characteristics of soils, topography, or surficial geology. Sensitive areas may be defined by focusing primarily on terrestrial or aquatic system features. Examples of datasets and GIS maps that can be used to locate ecosystem types are given in

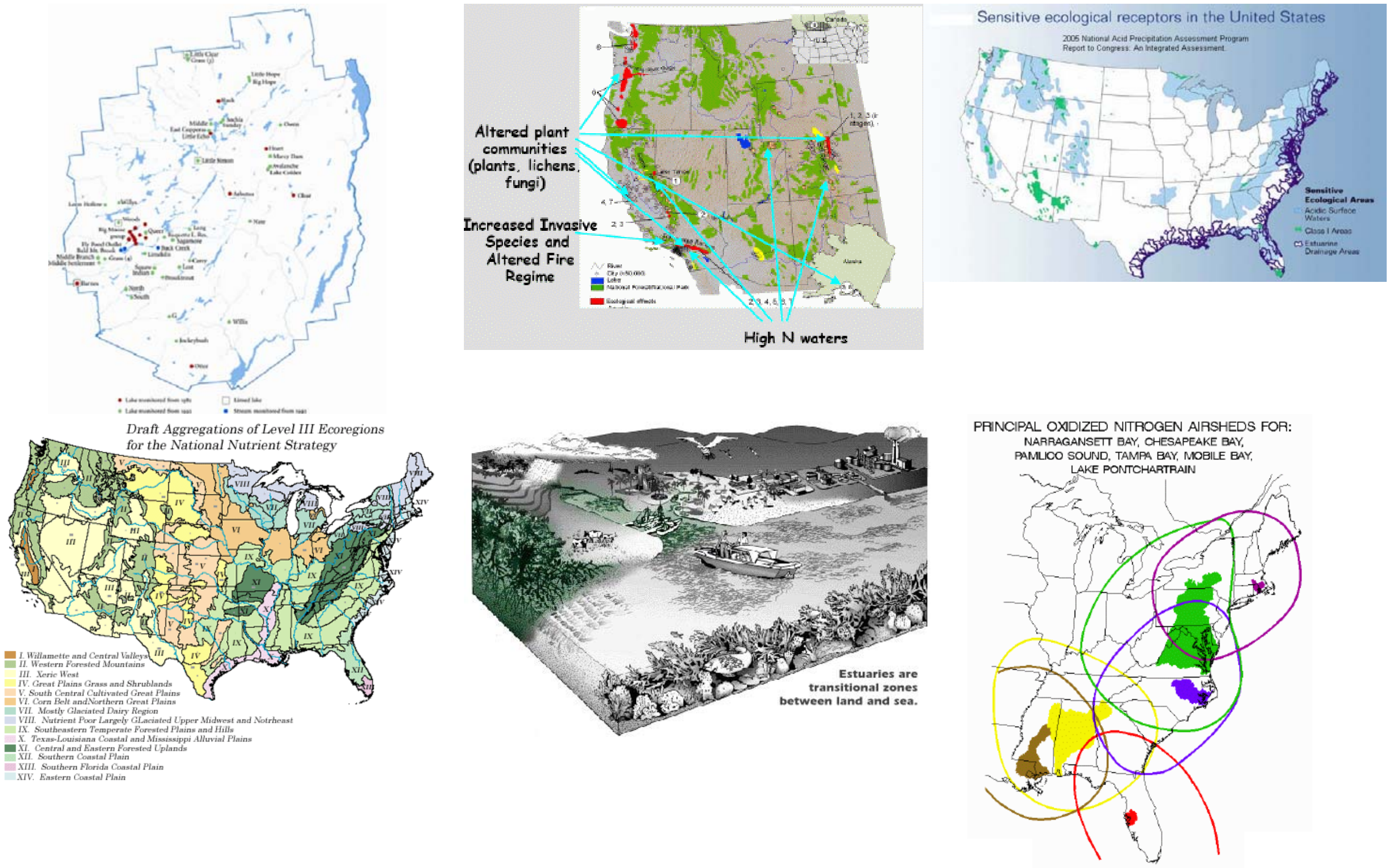


Figure 3-3. Documented biological, biogeochemical, and physiographic linkages.

Table 3-4. These materials may also be of value in identifying a list of nitrogen- and sulfur-sensitive ecosystem services and patterns and trends due to changes in reactive nitrogen and sulfur inputs (Section 3.7 describes this step).

Table 3-4. Example Datasets Planned for GIS Analysis

Ecosystem Type	Description	Effect of Interest	Indicator Parameter	Internet Link or Other Resource
Lakes, rivers, estuaries	Nutrient criteria for lakes, rivers, and estuaries	NE	TN for lakes and rivers	www.epa.gov/waterscience/criteria/nutrient/ecoregions
Lakes, rivers, estuaries	Acid sensitive waters of the United States	ACID	pH, ANC	epamap4.epa.gov/cmap/viewer.htm
All	National atlas of Class I areas	NE and ACID	All	www.nationalatlas.gov/mld/fedlanp.html
All	Ecoregions of the United States	NE and ACID	All	nationalatlas.gov/atlasftp.html#ecoomrp
All	Soil characterization data	NE and ACID	pH, ANC, exchangeable Ca^{2+}, dissolved organic content	www.ncgc.nrcs.usda.gov/products/datasets/statsgo/description.html
Lakes, rivers, estuaries	Total nitrogen deposition and other analytes	NE	NH_4^+	epamap4.epa.gov/cmap/viewer.htm
Forests and grasslands	U.S. vegetation	NE and ACID	N and S	ivm.cr.usgs.gov/products.php Also available from NationalAtlas.gov
Rivers, lakes, estuaries	Surface water quality		Many	www.epa.gov/storet
High-elevation–based lakes and rivers	U.S. elevation map	ACID	S	ESRI 8.3 data disks
All	Land cover	NE and ACID	N and S	www.mrlc.gov/mrlc2k_nlcd.asp
All	NADP grid files	NE and ACID	N and S	nadp.sws.uiuc.edu/isopleths/grids.asp

Note: NE = nutrient enrichment, ACID = acidification, TN = total nitrogen, S = sulfur, N = nitrogen

Recommended Mapping Layers and Models: Table 3-5 summarizes the current plan for GIS mapping layers and models to be applied in the risk/exposure assessment of targeted sensitive ecosystems. This plan is preliminary and will be reassessed as EPA proceeds in performing the seven steps for characterizing ecological effects.

3.3 Step 3 – Select Risk/Exposure Case Study Assessment Area Within a Sensitive Area

We intend to use the sensitive areas identified via Step 2 to select/delineate case study assessment areas for the risk/exposure assessment. Where case study or ecosystem-specific data are available, a subset of maps for the case study assessment area may be created. Complementary to these efforts, we may use a statistical cluster analysis to group ecosystem units into similar sets. Clustering ecosystems might reduce the number of locations that need to be modeled to adequately characterize the variability in ecosystem response to changes in nitrogen and sulfur deposition.

In selecting areas to assess ecological effects from air deposition, the SAB Ecological Effects Subcommittee (EES) suggests consideration of (1) clear quantifiable ecological effects due to air pollution, (2) the degree to which a significant component of ecological effects are attributable to air pollution, (3) the responsiveness of ecosystem services to changes in air pollution,(4) the cumulative impacts of multiple air pollutants, (5) the abundance of ecological effects and economic benefit cost analysis, and (6) the visibility to the public and value of resources at risk (U.S. EPA, 2005). While these recommendations were made in the context of a prospective cost-benefit analysis, many of these recommendations are sound in the context of our NAAQS risk analysis. The EES also provided some specific critiques and recommendations of specific potential case study assessment areas. We plan to evaluate their comments as part of our case study assessment area selection process. Among other points the EES emphasized are as follows:

- "Be cautious in any chosen assessment area when using surrogate sources to quantify ecological effects of air pollutants. For example, 'New' nitrogen (or mercury) derived from air pollutants is generally more bioavailable than 'old' nitrogen (or mercury). Moreover, ecosystems and associated organisms respond differently to different species and sources of nitrogen (and mercury)." (p. 6).

Table 3-5. Summary of Indicators, Mapping Layers, and Models for Targeted Ecosystems

Targeted Ecosystem Effect	Indicator(s)	Mapping Layers	Model(s)	Remarks
Terrestrial Nitrogen Enrichment	CEC C:N ratios Ca:Al ratios Air wet/dry deposition (corrected for throughfall using available data)	Forest soils from USFS Forest type from USFS Statsgo soils NLCD CMAQ (N) by HUC	MAGIC; PnET-BCG	
Aquatic Nitrogen Enrichment and Eutrophication	Nitrate and ammonia, total nitrogen (major reactive N species) Al toxicity data Chl-a (algal standing crop) anoxia/ hypoxia (primarily estuaries and tidal rivers) N loadings for sub-watersheds or larger basins and Estuarine Drainage Areas (EDAs) EPA NCCR WQ index and NOAA Estuarine-Coastal Eutrophication Index Diatom data for N-limited systems	STORET retrievals USGS NAWQA information USGS SPARROW information WQS Nutrient Criteria for rivers and lakes EPA NCCR and NOAA estuarine eutrophication indicators NOAA EDAs EPA/NOAA airsheds for major Atlantic and Gulf estuaries CMAQ (N) by HUC	USGS SPARROW PnET-BCG	
Aquatic Sulfur Enrichment (MeHg Focus)	MeHg (ambient) MeHg (tissue residues) Sulfur (ambient and sediments)	Devils Lake, WI, area	Limit to review of previous research	Examine studies for Devil's Lake, WI; also examine other literature from Mercury Report to Congress and recent work for areas in the northeastern United States
Terrestrial Acidification Due to Nitrogen and Sulfur	Soil ANC Soil pH CEC Inorganic Al Ca:Al ratio	Special areas (e.g., Class I areas, the Adirondack Mountains) CMAQ (N & S) by HUC Forest soils from USFS Statsgo soils USFS lichen USFS forest types	MAGIC; ILWAS; PnET-BGC	

Targeted Ecosystem Effect	Indicator(s)	Mapping Layers	Model(s)	Remarks
Aquatic Acidification Due to Nitrogen and Sulfur	ANC pH Al NO_3 and SO_4 fluxes or loadings	Acid-sensitive waters Select Class 1 areas EPA STORET USGS NWIS CMAQ (N & S) by HUC	MAGIC; PnET-BGC; SPARROW	

Note: CEC = cation exchange capacity, C:N = carbon:nitrogen, Ca:Al = calcium:aluminum, Chl-a = chlorophyll a, NCCR = National Coastal Condition Reports, WQ = water quality, NOAA = National Oceanic and Atmospheric Association, S = sulfur, USFS = U.S. Forest Service, NLCD = National Land Cover Data, HUC = hydrological unit, STORET = STOrage and RETrieval, USGS = U.S. Geological Survey, NAWQA = National Water Quality Assessment Program, WQS = water quality standards, NWIS = National Water Information System, ILWAS = Integrated Lake-Watershed Acidification Study.

- "It is important to choose [assessment areas] where atmospheric deposition itself can be distinguished from other sources contributing to ecological effects of interest. Thus, selection of an [appropriate assessment] area should be based not only on the type of ecosystem and its geographical location, but on the sources and types of air pollutants that impact it." (p. 7)
- "The EES encourages the EPA to consider sites in different regions with different resources at risk to help bring attention to the importance of ecosystem valuation." EPA could take advantage of the "opportunity to examine the effects of control of multiple pollutants individually and in combination." (p. 7)

The EES also provided a summary table listing potential case study areas for examining ecological benefits of reducing atmospheric deposition. This table is reproduced in **Table 3-6**. The ISA also recommended case study areas as candidates for risk/exposure assessments (**Table 3-7**). Special emphasis was given to the following:

- Adirondacks
- Shenandoah National Park
- Chesapeake Bay
- Alpine and subalpine areas of the Rocky Mountains.

We plan to consider these case study areas and any additional sites identified by the Step 2 mapping exercise in our selection of risk/exposure assessment areas. Options for selecting case study assessment areas include site-specific quantitative modeling analysis, qualitative analysis, cluster analysis, and review and summary of previous risk/exposure assessments. It is likely that since multiple risk/exposure assessments may be needed depending on the number of effects characterized, a combination of assessment methods will be used.

Table 3-6. SAB/EES Listing of Potential Assessment Areas for Evaluation of Benefits of Reductions in Atmospheric Deposition

Ecosystem/ Region	Main CAA Pollutant(s)	Percentage(s) Attributable to Atmospheric Deposition	Quantitative Ecological and Economic Information	EES Comments
Coastal				
Waquoit Bay	Nitrogen	30%	Yes	High priority. Higher loading from non-depositional sources may confound analysis.
Chesapeake Bay	Nitrogen	20–30%	Yes	High priority. Loading from diverse sources, particularly agricultural, may confound analysis.
Long Island Sound	Nitrogen; Mercury	Nitrogen = 23–35%; Mercury = ?	Yes	High priority. High nitrogen loading from wastewater treatment plants may confound analysis.
Everglades	Mercury	20–85%	Ecological = yes; Economic = uncertain	Medium priority. Reduction in atmospheric deposition has already resulted in decreased mercury burdens in fish and other biota.
Lake Michigan	Mercury	87%	Ecological = yes; Economic = lacking	Medium priority. Lack of quantitative economic data may restrict analysis.
Barnegat Bay	Nitrogen	50% total Direct deposition 30–39%	Yes	High priority. Direct linkage of ecological effects with atmospheric deposition; quantitative economic data exist.
Tampa Bay	Nitrogen; Mercury	Nitrogen = 25–30%	Yes	Medium priority. Examined in previous EPA efforts. Variability in loading data may confound analysis.
Gulf of Maine	Nitrogen	Low	?	Low priority. Linkage of nitrogen loadings and ecological impacts is not well established. Major source of nitrogen is open-ocean influx.
Casco Bay	Nitrogen; Mercury	Nitrogen = 30–40% Mercury = 84–92%	Yes	Medium priority. Good data on ecological and economic impacts are available.

Ecosystem/ Region	Main CAA Pollutant(s)	Percentage(s) Attributable to Atmospheric Deposition	Quantitative Ecological and Economic Information	EES Comments
Forested				
Adirondacks	Nitrogen; Sulfur; Mercury	Nearly 100%	Yes	High priority. Good quantitative ecological and economic data exist. Previous studies can be augmented readily.
Catskills	Nitrogen; Sulfur	Nearly 100%	Yes	Medium priority. Economic data may be lacking. Issues similar to the Adirondacks.
Southern Appalachian Mountains	Nitrogen; Sulfur	Nearly 100%	Yes	Medium priority. Economic data on fisheries are available. Issues similar to the Adirondacks.
Rocky Mountains	Nitrogen	Nearly 100%	Yes	Medium priority. Levels of nitrogen loading much lower than for northeastern locations. Economic data may be lacking.

Table 3-7. Potential Assessment Areas Identified in the Draft ISA (U.S. EPA, 2007b)

Area	Indicator	Detailed Indicator	Area Studies	Models	References in EPA, 2007b	
Adirondacks	Aquatic Nutrient Enrichment; Terrestrial Nutrient Enrichment; Mercury Methylation		PIRLA I and II; Adirondack Lakes Survey; Episodic Response Project; EMAP	MAGIC; PnET-BGC	Baker and Laflen, 1983; Baker et al., 1990b; Baker et al., 1990c; Baker et al., 1996; Benoit et al., 2003; Chen and Driscoll, 2004; Confer et al., 1983; Cumming et al., 1992; Driscoll et al., 1987a; Driscoll et al., 1991; Driscoll et al., 1998; Driscoll et al., 2001a; Driscoll et al., 2001b; Driscoll et al., 2003b; Driscoll et al., 2003c; Driscoll et al., 2007a; Driscoll et al., 2007b; Evers et al., 2007; GAO, 2000; Havens et al., 1993; Ito et al., 2002; Johnson et al., 1994b; Landers et al., 1988; Lawrence et al., 2007; NAPAP, 1998; Siegfried et al., 1989; U.S. EPA, 2003; Sullivan et al., 1990; Sullivan et al., 2006a; Sullivan et al., 2006b; U.S. Environmental Protection Agency, 1995b; Van Sickle et al., 1996; Whittier et al., 2002; Wigington et al., 1996; Zhai et al., 2007	ISA
	Aquatic Acidification			ILWAS	Gherini et al., 1985	Lit. Search
Shenandoah National Park	Aquatic Acidification; Terrestrial Nutrient Enrichment		Shenandoah Watershed Study; Fish in Sensitive Habitats (FISH) study	MAGIC	Baker and Christensen, 1991; Baker et al., 1990b; Bulger et al., 1999; Bulger et al., 2000; Cosby et al., 2006; Dennis and Bulger, 1995; Dennis et al., 1995; Deviney et al., 2006; Eshleman and Hyer, 2000; Eshleman et al., 1995; Eshleman et al., 1998; Galloway et al., 1983; Hyer et al., 1995; MacAvoy and Bulger, 1995; Molot et al., 1989; Schofield and Driscoll, 1987; Sullivan et al., 2003; Sullivan et al., 2007a; Webb et al., 1995	ISA

Area	Indicator	Detailed Indicator	Area Studies	Models	References in EPA, 2007b	
Chesapeake Bay	Aquatic Nutrient Enrichment; Aquatic Nitrogen Limited Eutrophication				Bricker et al., 1999; Bricker et al., 2007; Boesch et al., 2001; Boyer et al., 2002; Boyer and Howarth, 2002; Cooper and Brush, 1991; Fisher and Oppenheimer, 1991; Harding and Perry, 1997; Howarth, 2007; Kemp et al., 1983; Malone, 1991, 1992; Officer et al., 1984; Orth and Moore, 1984; Twilley et al., 1985	ISA
Alpine and Subalpine Communities of the Eastern Slope of the Rocky Mountains in Colorado	Aquatic Nutrient Enrichment; Terrestrial Nutrient Enrichment	biomass production; NO_3 leaching; species richness			Baron et al., 1994; Baron et al., 2000; Baron, 2006; Bowman, 2000; Bowman and Steltzer, 1998; Bowman et al., 1993; Bowman et al., 1995; Bowman et al., 2006; Burns, 2004; Fenn et al., 2003a; Fisk et al., 1998; Korb and Ranker, 2001; Rueth et al., 2003; Seastedt and Vaccaro, 2001; Sherrod and Seastedt, 2001; Steltzer and Bowman, 1998; Suding et al., 2006; Williams and Tonnessen, 2000; Williams et al.,1996a; Wolfe et al.,2001	ISA
Beartooth Mountain, Wyoming	Aquatic Nutrient Enrichment	algae composition switch			Saros et al., 2003	ISA
Fernow Experimental Forest near Parsons, West Virginia	Terrestrial Nutrient Enrichment	forest growth			Adams et al., 1997, 2000; DeWalle et al., 2006; Edwards and Helvey, 1991; Gilliam et al., 2006; Peterjohn, 1996	ISA
Uinta Mountains of Utah and the Bighorn Mountains of central Wyoming	Aquatic Acidification	lake NO_3 concentrations	Western Lakes Survey		U.S. EPA, 1987	ISA
Pamlico estuary in North Carolina	Aquatic Nitrogen Limited Eutrophication	hypoxia; phytoplankton bloom			Paerl et al., 1998	ISA

Area	Indicator	Detailed Indicator	Area Studies	Models	References in EPA, 2007b	
Bear Brook, Maine	Aquatic Acidification; Terrestrial Nutrient Enrichment	sugar maple; red spruce			Elvir et al., 2003	ISA
Harvard Forest	Terrestrial Nutrient Enrichment	forest growth—species			Magill et al., 2004; Magill, 2004	ISA
Southern California	Terrestrial Nutrient Enrichment	forest growth—species; coastal sage scrub			Fenn et al., 1996, 2003a; Takemoto et al., 2001	ISA
Jasper Ridge Biological Preserve in California	Terrestrial Nutrient Enrichment	grasslands			Zavaleta et al., 2003	ISA
Allegheny Mountains of West Virginia	Aquatic Acidification	high streamwater or lake NO_3 concentrations			Gilliam et al., 1996	ISA
Catskill Mountains of New York	Aquatic Acidification	high streamwater or lake NO_3 concentrations			Murdoch and Stoddard, 1992; Stoddard, 1994	ISA
Great Smoky Mountains in Tennessee	Aquatic Acidification	high streamwater or lake NO_3 concentrations			Cook et al., 1994	ISA
Loch Vale, Colorado	Terrestrial Nutrient Enrichment	old-spruce growth			Rueth et al., 2003	ISA
Rocky Mountain National Park, Colorado	Terrestrial Nutrient Enrichment	tundra composition switch				
	Aquatic Nutrient Enrichment	diatom shifts			Interlandi and Kilham, 1998	ISA

Area	Indicator	Detailed Indicator	Area Studies	Models	References in EPA, 2007b	
Rocky Mountain National Park, Colorado	Aquatic Acidification	subalpine lakes		MAGIC	Sullivan et al., 2005	Lit. Search
Lake Tahoe, California	Aquatic Nutrient Enrichment	primary productivity; chlorophyll a			Goldman, 1988; Jassby et al., 1994	ISA
Little Rock Lake, Wisconsin	Aquatic Sulfur-Enhanced Mercury Methylation	bioaccumulation of Hg in freshwater fish			Hrabik and Watras, 2002; Watras and Frost, 1989; Watras et al., 2006	ISA
Southern Appalachians	Aquatic Acidification	Streams		MAGIC	Sullivan et al., 2004	Lit. Search
Hubbard Brook, New Hampshire	Terrestrial Acidification; Aquatic Acidification	forest ecosystem; soils; streams	many studies for decades	PnET-BGC	Gbondo-Tugbawa and Driscoll, 2002; Gbondo-Tugbawa et al., 2002	Lit. Search

3.4 Step 4 – Evaluate Current Loads and Effects to Case Study Assessment Area Including Ecosystem Services

This step involves evaluating the deposition and terrestrial and aquatic fate and transport of nitrogen and sulfur, as well as the ecological effects, and their subsequent effect on ecosystem services, resulting from exposure to certain levels of nitrogen and sulfur. Depending on the adequacy and abundance of data for areas, the evaluation may entail computer modeling, statistical analysis, or qualitative analysis.

3.4.1 Assess Data Availability and Adequacy

Determine Which Indicators Have Monitoring Data and/or Modeling data Available for Analysis: The ISA contains a review of recent (2000 to present) monitoring programs for targeted ecosystem effects and associated indicators. We propose to use ANC as the first choice of acidification indicators. However, we may consider other indicators if we determine that data are insufficient or other factors demonstrate a need to change indicators. Nitrogen and sulfur enrichment have both terrestrial and aquatic impacts, making the selection of indicators more challenging. Currently, we have narrowed the list of indicators for additional evaluation to the following:

- Wet and dry nitrate and ammonia deposition
- Nitrate, ammonia, organic nitrogen throughfall deposition for terrestrial ecosystems
- Total nitrogen or constituent species' loadings and fluxes to receiving waters from runoff, air, discharges, and groundwater inputs
- Algal standing crops and anoxia/hypoxia for aquatic systems.

Environmental monitoring data and programs will be used to detect long- and short-term effects of nitrogen and sulfur deposition. Therefore, we will consider the conditions of the monitoring programs supplying data for this assessment; for example, nationally sponsored, long-term studies versus short-term academic research.

The ISA also reports literature available to assess impacts on ecosystem services that can be used to more fully describe the importance of effects on services that are important to the public.

Evaluate the Adequacy of Spatio-temporal Data and the Statistical Adequacy of Available Data: We plan to evaluate the spatial adequacy of available monitoring data including GIS mapping of documented data to identify any meaningful spatial gaps. For each ecosystem effect, we plan to determine if there is a temporal dimension to exposure. That is, if effects “lag” behind exposure at different scales of time, we plan to develop an approach to address those time-steps (e.g., measure effects on a daily or annual scale), and we plan to seek and assess data available for candidate indicators from both monitoring and modeling data that address the temporal dimensions required.

We intend to review and determine the best indicators for acidification and nitrogen and sulfur enrichment for each targeted ecosystem effect. To accomplish this, we plan to identify spatial and temporal gaps, document uncertainties and limitations raised in the research literature, and raise any additional points about uncertainty identifiable through either statistical analysis or qualitative evaluation (depending on the form of information available).

Resolve Gaps and Disadvantages (e.g., integrate datasets and/or models; interpolate): The location and type of ecosystem effects resulting from nitrogen and sulfur deposition do not lend themselves to a traditional large-scale risk/exposure assessment. This is due to both the isolated or regional effects observed and the potential lag in time for observed effects. Therefore, it will likely be necessary to resolve spatial and temporal gaps in data needed to perform a risk/exposure assessment. We propose to resolve gaps through a combination of integrating datasets, modeling, and interpolation/extrapolation. We plan to use CMAQ output data at 12 km grids for regions of interest, and an appropriate multimedia model to address spatial data needs for terrestrial and aquatic exposure assessments for the targeted ecosystem effect. Additional information on CMAQ and multimedia models is presented in Appendices C and D, respectively.

3.4.2 Compute Loading and Exposure for Each Ecosystem Effect:

We plan to use current data and models to analyze reactive nitrogen and sulfur loads and exposures. Major categories of loading data include the following:

- Atmospheric deposition across the landscape (available from CMAQ modeling for 2002 for 8-digit hydrological units [HUCs] and a 12-km grid) (**Figure 3-4** presents past applications of CMAQ modeling to support the evaluation of National Acid Precipitation Assessment Program [NAPAP]–monitored acidification effects in the United States.)

- Atmospheric deposition monitoring data (National Atmospheric Deposition Program [NADP] and Clean Air Status and Trends Network [CASTNET])
- National Pollutant Discharge Elimination System (NPDES) point source discharge data
- Agricultural runoff modeling
- Urban non-point source runoff modeling
- Other urban or rural loading sources (e.g., onsite septic system modeling).

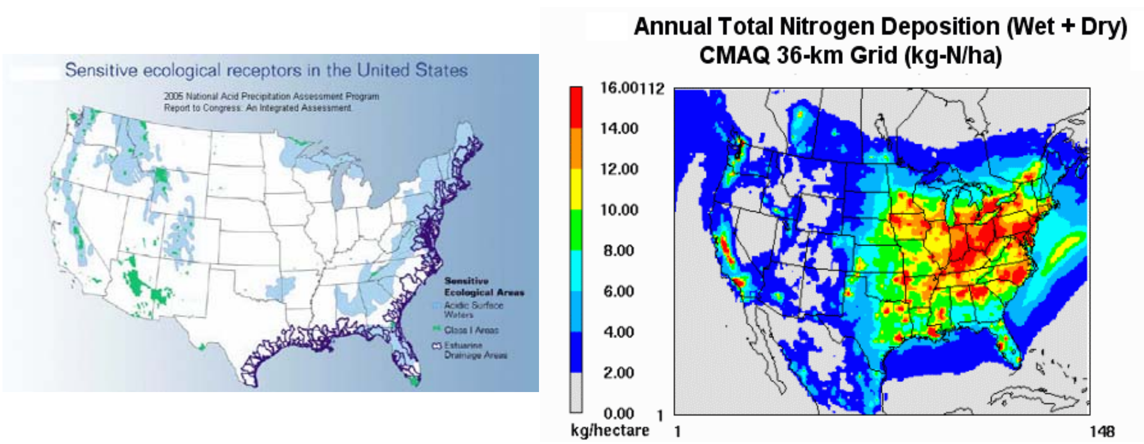


Figure 3-4. Comparison of NAPAP-documented acid-sensitive ecoregions to CMAQ-modeled nitrogen deposition.

CMAQ Deposition Modeling: It is necessary to understand the role of receptor surfaces (i.e., land use/land cover) influencing atmospheric deposition. Receptor surfaces affect the dry deposition of gases and particles. Dry deposition can contribute significantly to total deposition (in many locations and for many chemical species >50% of total deposition). CMAQ modeling should account for the role of deposition due to both atmospheric conditions and land cover. We plan to use land cover datasets, such as the National Atlas, to support our analysis of the role of land cover in targeted ecosystem effects. Appendix C provides a more extensive description of CMAQ modeling plans for this risk/exposure assessment.

Multimedia Modeling: The ISA (U.S. EPA, 2007e) provides a survey of multimedia models, and a summary of the ISA review is in Appendix D. The recommended models for the risk/exposure assessment are presented here. The two distinct environmental effects of nitrogen and sulfur deposition for which indicators can be defined and models identified based on past applications, geographic applicability, and use of atmospheric inputs are acidification and nitrogen and sulfur enrichment.

To provide more detail for the models, those that readily accept atmospheric deposition inputs of the same nature as CMAQ output (flux in mass per area per time) are presented in bold. Those that accept atmospheric concentration data (mass per volume) are presented in italics. Although these classifications note whether a model accepts atmospheric deposition input, they do not specify whether temporal and spatial resolutions will match between the atmospheric and ecosystem models.

Acidification

As explained in Appendix A, acidification is an environmental effect in which acid precipitation lowers the natural pH of waterbodies and/or damages terrestrial ecosystems. The key indicators of acidification likely to be considered in any modeling efforts to determine the effects of acidification on an ecosystem include both chemical measurements and ecological/biological indices and factors. The biological indicators of acidification can be found in both terrestrial and aquatic systems; however, models are much less developed for simulating or estimating measures of the biological indicators on land. The chemical indicators of acidity are much more likely to be included in a model.

Both **MAGIC** and ***WARMF***[1] account for several of the chemical indicators of acidification for both terrestrial and aquatic systems. Either of these models could be used for an acidification analysis, although ***WARMF*** contains a more robust analysis with higher-level processes, including biological processes, and a GUI.

The ILWAS and WARMS models both provide for acidification indicators but have limited applications outside of their development areas of Illinois and Ontario, respectively. ILWAS is a highly parameterized model.

The TMDB/IBIS model does not include the minerals as its state variables but does simulate nitrogen and carbon, as well as vegetation, which could be considered a measure of forest health. This model can be used on a global scale.

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[1] WARMF currently accepts atmospheric deposition in concentration form; however, EPRI and Systech Engineering, Inc., the developers of WARMF, have recently undertaken a project to modify WARMF to accept depositional fluxes such as those output by CMAQ.

Several of the terrestrial/watershed models could be used to provide input loads to a receiving-water model for acidification studies. The choice of model would depend on the time-step needed and the geographic location of analysis. Applicable terrestrial/watershed models include **PnET-BGC**, DayCent, Century 4/5, *NuCM*, and **SAFE**.

Our current intention is to run MAGIC for a case study area to look at changes in ANC. The other models may be considered if further analysis would prove useful.

Nitrogen Enrichment

Indicators of nitrogen enrichment are addressed in detail in Section 3.1.1. Atmospheric deposition has been shown to increase nitrogen enrichment in Atlantic Coast estuaries and is now of concern in high-altitude, alpine lakes. Because the key indicators for nitrogen enrichment depend on measurements of nitrogen, there are many models available to estimate the ecosystem effects, but special attention should be paid to how these models simulate the nitrogen cycle and whether biological processes, which are vital to the cycle, are included. Nitrogen, either total or speciated, is probably the most highly modeled chemical parameter in ecosystems. Both receiving-water and terrestrial/watershed models may be used in these analyses. Additionally, it is possible to use a combination of models so that a watershed model provides input loadings to the receiving water model.

The following receiving-water models all estimate speciated nitrogen (e.g., ammonia, nitrate, organic, or total) and can be used in most geographic locations: *AQUATOX*, **QUAL2K**, **WASP**, and the **CE-QUAL** family of models.

The THMB/IBIS model simulates the nitrogen cycle, but has limited geographic applicability for small sites. It is more applicable for large-scale simulations.

HSPF, SWAT[2], and ***WARMF*** simulate the nitrogen cycle across land and water; however, SWAT contains a more simplified approach to the nitrogen cycle in reservoirs and lakes. **HSPF** accepts the atmospheric flux of nitrogen that is output by CMAQ. While SWAT currently only considers a state nitrate concentration in wet deposition, ***WARMF*** currently

[2] SWAT currently accepts only a static input nitrate concentration in precipitation. An untested concept to utilize SWAT's fertilizer management function to simulate atmospheric deposition may provide a work-around function.

accepts atmospheric concentrations of NO_x, ammonia, and nitrate in both wet and dry atmospheric conditions.

Nitrogen enrichment, more than acidification, may be measured across the terrestrial landscape, in addition to waterbodies. Many of the terrestrial/watershed models may be used for nitrogen enrichment analyses, with the choice depending on time-step, geographic location, nitrogen species desired, and simulation of the nitrogen cycle within the model. For daily simulations, possible models include: DayCent, DNDC, *DRAINMOD-N II*, *EPIC/APEX*, GLEAMS, **INCA**, **RHESSys**, and **GT/MEL**. For longer-term simulations (monthly, annual), the following models may be used: Century, **MAGIC**, **MERLIN**, **PnET-BGC**, ReNuMa, and *SPARROW*.

Approach for Selecting Geographic Regions to Model: Selection of geographic regions of the United States to model will depend on a number of factors:

- Observed data that are indicative of the ecosystem and response of interest or observed characteristics of an ecosystem that implies it has the potential to respond
- Availability of a model capable of analyzing the region of interest
- Availability of model input data for the region.

It may be possible to explore adapting an existing model to an untested geographic area or using synthetic or surrogate data if there is a geographic region of particular interest with insufficient model input data. In contrast, if there are multiple geographic regions suitable for modeling, we intend to examine the feasibility of clustering the regions, increasing the scale of the modeling to capture the regions, or selecting the one region with the best potential to provide valid and representative data on ecosystem response.

Assess Uncertainty in Loading and Exposure Computations: The risk/exposure assessment will need to account for the fact that current runoff models vary in resolution. Runoff and other fate and transport models should be selected to adequately address the differing complexities of the identified sensitive ecosystems. To the extent possible, the analyses should try to include all reactive nitrogen species, recognizing that inventories may need improving. Most likely, the assessments will initially be a snapshot of the loadings and exposures with more dynamic assessments to follow in future reviews. Where possible, the focus will be to develop the best available numeric measures of the loading contribution from all major sources, so that the contributions from air deposition sources can be analyzed relative to all other major anthropogenic or natural sources for sensitive ecosystems across the country.

3.5 Step 5 – Where Feasible, Scale-Up Case Study Assessment Area Findings to Sensitive Areas

Several approaches can be applied to take the results of analyses for specific case study assessment areas and relate these findings to more spatially extensive sensitive areas. For instance, analyses using MAGIC taking advantage of good quality Temporally Integrated Monitoring of Ecosystems (TIME) and Long-Term Monitoring (LTM) data sets may be scaled up to several extensive ecological and physiological provinces such as the Adirondacks, the Appalachian Plateau (primarily in Pennsylvania), and the Ridge & Blue Ridge Provinces (largely in the Shenandoah National Park and along the Shenandoah Parkway). Where the TIME/LTM datasets have previously been applied to define the extent of sensitive areas, scaling up the new analyses to the same previously used larger sensitive areas may be readily justified. Where the case study assessment areas are defined using clusters of sampling sites, more sophisticated GIS-based spatial interpolation techniques may be applied where the case study assessment area sites are related to bounding polygons reflecting standard ecoregion delineation systems (e.g., the EPA Omernik Level II ecoregions or the aggregated Omernik ecoregions developed for EPA criteria for numeric nutrient criteria for lakes and streams). Statistical cluster analysis techniques may be applied to provide a means for objectively grouping localized case study assessment areas into larger sensitive areas. For larger case study assessment areas such as the Chesapeake Bay system, available national indicator products such as the USGS SPARROW models for HUC8 (sub-basin) watersheds, or EPA and NOAA estuary eutrophication indicators may be considered as tools to document similarities and differences for major estuarine and near coastal aquatic systems, taking into account the relative importance of atmospheric deposition of pollutants such as nitrogen to sources within estuarine drainage areas related to point source, urban, or rural nutrient loadings.

A qualitative or semi-quantitative characterization of these larger sensitive areas may be developed, depending on data availability and time constraints. Where feasible, we plan to also discuss the effect on related ecosystem services of these sensitive areas.

3.6 Step 6 – Assess the Current Ecological Conditions for Those Sensitive Areas

3.6.1 Calculate Response Curves for Each Indicator

If feasible, we intend to generate indicator response curves from the multimedia model runs. The response curves can supply useful information about the degree of impairment and sensitivity of the system.

3.6.2 Calculate Desired Exposure Endpoint

Once the sensitive ecosystems response curves are generated, it may be possible to calculate a desired exposure endpoint. This requires similar information and steps required for a critical load analysis. Critical load analyses may be considered, where appropriate, in identifying ways to select endpoints in terms of ecosystem responses relative to atmospheric levels (concentrations or loadings) of nitrogen and sulfur. A detailed description of critical loads and its use as an analysis tool are presented in the ISA (U.S. EPA, 2007b).

3.6.3 Develop GIS Maps and Overlay Loading and Indicators That Are Representative of Harmful Effects

For each targeted ecosystem effect, we plan to create GIS maps from the modeling and monitoring data generated/compiled for selected indicators. Then, a second data layer can be created to represent the level of exposure at which harmful effects based on the indicator, and these data layers can be combined to facilitate mapping.

3.6.4 Using GIS Mapping, Compute the Extent That Loading Is Greater Than or Less Than the Harmful Effect Level (a.k.a. endpoint)

We plan to use GIS to compute the difference in exposure relative to the response-based harmful effect level. We recognize that the loading and exposure may be less than or greater than the findings of the exposure response research. Based on our findings, we plan to determine if a particular geographic region requires additional analysis or if we are able to proceed to Step 7.

3.6.5 Assess Ecosystem Responses via Ecosystem Services and Valuation

One component of an integrated assessment of risks to the public welfare involves determining what level of ecosystem response translates into an effect that could reasonably be considered adverse to the public welfare. There are a number of ways to do this, including the following:

- Direct measures of quantities that are of known value to the public (e.g., numbers of endangered species)
- Translation of ecosystem attributes into measures of ecosystem services, which can then be quantified
- Direct economic valuation of ecosystem functions and services, including use and nonuse values (values that do not require an individual’s direct use of an ecosystem—for example, the value of preserving an endangered species habitat, even though that individual will ever see that species in the wild)
- Direct nonmonetary valuation of ecosystem functions based on enumeration of preferences using nonmonetized indices of preferences.

The specific methods used to evaluate adversity will depend on the availability of data and methods for the indicators of interest related to acidification and nutrient enrichment and on an assessment of the appropriateness of each type of quantification in comparing different levels and forms of the standards. In our initial assessment of the available data, given the timeframe for this NAAQS review, we have determined that the most useful approaches will be those focusing on quantifying the link between changes in ecosystem indicators and ecosystem services. Linking these changes in ecosystem services to changes in economic values will likely be beyond the scope of the current assessment. A brief discussion is provided in Appendix E on how valuation approaches might be used to combine and/or compare changes across ecosystem services.

The EPA SAB Committee on Valuing the Protection of Ecological Systems and Services (SAB CVPESS) has recently drafted major recommendations for improving ecological benefits assessment to facilitate consideration of ecosystem benefits in the decision-making process. This SAB panel recommends taking “a more comprehensive approach to assessing, valuing, and reporting on the ecological benefits of its actions” (SAB CVPESS, 2007). The conceptual model described in **Figure 2-5** that links atmospheric concentrations and deposition to ecosystem effects and biologically relevant indicators, and tying these to ecosystem services and valuation is supported by the SAB panel. One of the critical gaps identified by the SAB panel is “identifying how the biophysical effects of an action on an ecosystem will in turn impact the ecosystem services of importance to the public” (SAB CVPESS, 2007).

In broad terms, the most important information gap in this review is an incomplete understanding of how hydrological and biogeochemical processes (e.g., cycling of H_2O, N, C, P) interact to control the response of ecosystem services to NO_x and SO_x and other forcing variables (e.g., climate). A major consequence of these interactions is that ecosystem services tend to be linked or "bundled," so that actions taken to improve one may result in an improvement or deterioration of other services. Thus, when ecosystem services are quantified and their ecological response functions to NO_x and SO_x are modeled, it is imperative that the entire bundle of services be evaluated, and that the linkages and tradeoffs among ecosystem services be included in the quantification (i.e., ecological tradeoff functions [ETFs]). The key feature distinguishing the services from the underlying ecosystem function or processes is the explicit involvement of beneficiaries, so consideration of which ecosystem services to target in assessment activities involves consideration of the relative demand for the service, its spatial distribution, and its magnitude.

Ecological Response Functions (ERFs) and Ecological Tradeoff Functions (ETFs) can be developed to quantify the response of a service to changes in NO_x/SO_x concentrations and the tradeoffs between different services given these ambient air quality drivers. Many of the services of importance in assessing risk of NO_x and SO_x have not been quantified (i.e., developed ERFs and ETFs) or adequately scaled for a comprehensive evaluation of the benefits of these services. Such research is being planned, and will, no doubt, contribute to future NO_x/SO_x assessments. Therefore, at this time, data mining will be central to developing at least a preliminary assessment of potential impacts of NO_x/SO_x deposition and acidity on ecosystem services. In the current plan, process-based models are being considered to be used to (1) synthesize/link the suite of ERFs and ETFs and (2) generate maps and summaries of ecosystem services and tradeoffs in response to current and future ambient air indicators for NO_x and SO_x. The collection of response and tradeoff functions will aid in the valuation of the services at risk to these criteria pollutants where possible.

A risk/exposure assessment approach using the most relevant and best available data on deposition, acidity, and measured effects will benefit from spatial and temporal mapping. In preparation for the NO_x/SO_x risk/exposure assessment, a suggested series of mapping exercises would proceed in the following order:

1. Identify areas/regions of the country receiving high levels of NO_x/SO_x deposition and acidity impacts.
2. In those regions, identify ecosystems sensitive to elevated levels of nitrogen and sulfur, using some common selection criteria.
3. In those sensitive ecosystems, ask what ecosystem services are expected to be prevalent and "valued" (i.e., some subset of all potential services).
4. In those areas, identify what data are available to develop ERFs and ETFs, at least to a qualitative degree that would enable production of spatial and temporal maps to identify different degrees of protection that would exist under alternative secondary NAAQS. Chan et al., (2006) produced such maps for several ecosystem services in the Central Coast ecoregion of California. The linkage and comparison of multiple ecosystem services in the region would provide information for consideration of tradeoff value of one service versus another.

Both economic and biophysical valuation can be considered in determining adverse effects and can be used in determining benefits of protection. As noted above, valuation is a useful way to compare disparate ecosystem impacts, and it is a potentially important component in the risk characterization phase of the risk assessment. As a result, an additional step in the above process is to identify potentially relevant economic valuation studies for ecosystem services and map the location of these studies relative to the available data on ecosystem services. Note that while we are considering all potential methods for obtaining ecosystem valuation estimates, the current focus of this plan is to use literature-based estimates of ecosystem service values previously identified in the ISA that can be applied in a valuation transfer approach. Realization of concepts like the conjoint analysis approach would require time and resources, which are not available for the current analysis, but which should be considered for future reviews.

3.7 Step 7 – Assess Alternative Levels of Protection Under Different Scenarios of Deposition From Ambient Sources

A secondary NAAQS standard, while national in scope, might have a form that allows for consideration of regional heterogeneity in ecosystem sensitivity and heterogeneity in atmospheric composition of nitrogen and sulfur deposition either alone or combined. Such a form should be designed to provide for the adequate protection of sensitive ecosystems and to allow flexibility for those ecosystems with more ecological resilience. Assessing alternative levels of protection with different loadings scenarios under different forms of the standards will be an important step in this process.

We intend the risk/exposure assessment to characterize exposure and ecological effects associated with current levels of nitrogen and sulfur deposition as described in this Scope and Methods Plan. However, the NAAQS are based on ambient concentrations of NO_x and SO_x in the atmosphere, and, therefore, additional analyses are required to move from deposition-based risk estimates to policy-relevant ambient indicators. The policy assessment will build upon the current conditions risk/exposure assessment to develop these policy-relevant ambient indicators (**Figure 3-5**). This requires synthesizing the ecosystem responses, biological indicators, and ecosystem effects related to deposition loadings and translating those loadings back to their corresponding ambient air conditions.

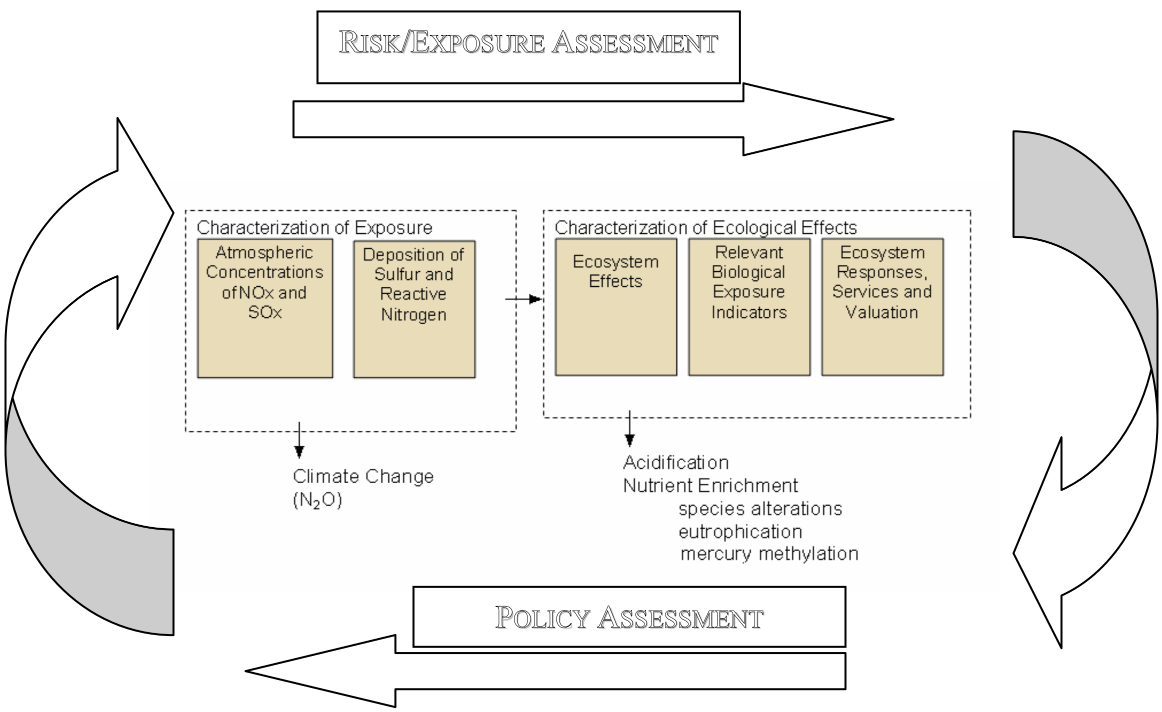

Figure 3-5. Risk/policy assessment paradigm for deposition-related ecological risks.

After the current conditions risk/exposure assessment develops an understanding of baseline conditions, it will be necessary to define the amounts of risk under varying levels and forms of the standards. The risk/exposure assessment intends to establish whether current levels are causing effects, which ecosystems are most sensitive to those effects, and the magnitude of the risks existing at current levels. After this baseline is established, modeling techniques can be used to change the ambient level and examine how the corresponding exposures and risks change. In this way, dose-response curves can be generated that establish the range of impacts and effects associated with nitrogen and sulfur inputs. For example, suppose at a particular level of loading (X), a negative effect is observed (Y). This analysis can be used to establish what X is equivalent to in terms of ambient concentrations, so that Y can be reduced or avoided. Several iterations of this process may be needed to examine different forms and levels of the standard(s) and determine if the form(s) of the standard(s) has an impact on the risk associated with that form.

Some of the issues associated with this type of analysis include:

- What adverse effects are we trying to protect against?
- If total nitrogen is the relevant biological indicator, what is the relative contribution of oxidized versus reduced forms of nitrogen, and what is the ambient contribution compared to other sources?
- Do these effects occur due to different ambient levels and/or forms of nitrogen and sulfur?
- How should alternative levels be selected (i.e., via a dose response curve, based on threshold events, or is another method more appropriate)?
- What are the correct temporal and spatial scales for the level of the ambient air indicator and how do they relate to the temporal and spatial scales of the deposition indicator?
- What are the associated uncertainties?

In determining the appropriate level and form of a standard, we plan to evaluate alternative levels and forms of the standard and evaluate the ecological risks associated with those levels and forms. Due to limits in data, modeling, and time, we are not able to conduct a national assessment. Instead, we plan to extend the assessment area analysis approach used in the current conditions analysis. Our initial thoughts on evaluating the appropriate level and form of the standard using the assessment area analyses approach include the following:

1. Identify the relative contribution of loadings associated with atmospheric deposition of nitrogen and sulfur.
2. Identify the most critical impacts from nitrogen and/or sulfur loadings (i.e., acidification, nutrient enrichment, or eutrophication).
3. Identify the contribution to atmospheric loadings from total reactive nitrogen, NO_x, and $SO_{x.}$
4. Identify the biogeochemical indicators/resources of concern in the assessment area and the ecosystem services associated with those indicators.
    a. Determine the ecosystem service effects associated with the most-critical impacts.
    b. Bundle ecosystem services to find common metrics for comparison across locations.
5. Define the exposure-response (loading-response) functions (ERFs) for the ecological indicators of concern.
6. Estimate the loadings/exposures associated with current and alternative levels of the NO_x and SO_x standards (using CMAQ modeling).
    a. Analyze the relationships between NO_x, SO_x, and other reactive forms of nitrogen.
    b. Assess the impacts of meteorological variability on these relationships.
7. Estimate the ecosystem impacts associated with estimated loadings.
8. Convert estimates of individual ecosystem risks to common units using
    a. economic valuation based on benefits transfer from existing literature estimates
    b. biogeochemical equivalents using ecological tradeoff functions (ETFs).
9. Combine individual risk estimates to produce overall impact estimates.

Results from individual assessments can be evaluated relative to the national maps of ecosystem types and sensitivities. This analysis may characterize how well the assessment areas represent overall national ecosystem types and sensitivities, and how similar impacts of alternative standards might be across ecosystems throughout the country. The output of the quantitative assessment and the science assessment feed the policy assessment, so that it can examine a range of alternative standards applicable for the agency to consider. The following are questions to address in identifying specific elements of standards:

- How do alternative levels and forms of the standards relate to a given exposure metric?
- What are the appropriate averaging times for alternative levels and forms of the standards?
- What alternative levels of the standards should be considered?
- Should there be alternative levels of the standards (i.e., individual NO_x and SO_x standards or a combined NO_x/SO_x standard)?
- Do the ambient air indicator forms allow for site-specific protection while maintaining national consistency?
- Does the ambient air indicator adequately account for the effects of total reactive nitrogen?
- Does the form of the standard have an impact on the risk?

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APPENDIX A

IDENTIFICATION OF SENSITIVE ECOSYSTEMS

A.1 Targeted Ecosystem Effects

One of the central issues in this secondary NAAQS review is determining which ecosystems are sensitive to nitrogen and sulfur deposition and their degree of sensitivity compared to one another. Characteristics to determine sensitivity may include, but are not limited to, (1) potential nitrogen and sulfur retention rates; (2) potential nitrogen and sulfur uptake rates, which might include vegetative uptake, potential denitrification, and potential mobilization of nitrogen and sulfur; (3) potential residence time based on local hydrology (precipitation rates, conductivity) and geology (bedrock type, pervious surfaces, soil properties); and (4) total supply of nitrogen and sulfur, including current and historical atmospheric deposition and other nonatmospheric sources (e.g., applied fertilizers, sewer leaks, point sources). Other ecosystem-specific characteristics that may help assess sensitivities include threatened and endangered species data where available, land cover, land-use type (including Class I, National Park, and Fish and Wildlife Refuges and National Wilderness areas), species of community shifts (or invasive species), and baseline nitrogen and sulfur loading estimates. Where ecosystem-specific data are available, a subset of maps for the study region may be created. Complementary to these efforts, we may use statistical cluster analysis to group ecosystem units into similar sets. By clustering ecosystems, we might reduce the number of locations that need to be modeled to adequately characterize the variability in ecosystem response to changes in nitrogen and sulfur deposition.

These types of analyses may aid in determining whether area-based risk/exposure assessments are appropriate for looking at nitrogen and sulfur effects on various ecosystems and geographic regions as a means of extrapolating these impacts to characterize the entire country. For those areas where data are available, watershed models (e.g., MAGIC, PnET-BGC, DayCent-chem) may be useful for evaluating the emission-deposition-ecosystem response linkage.

These are the main questions the risk/exposure assessment will address:

- If we can identify appropriate biologically relevant indicators, can we establish a link between deposition of NO_x (and/or reactive nitrogen), SO_x, and ecosystem response?

- What ecosystem services are associated with changes in emissions?

Ecosystem response to nitrogen and sulfur deposition varies across the landscape, depending on the local physical, chemical, and biological characteristics. Atmospheric deposition effects range from species alterations (due to nutrient enrichment) to anthropogenic eutrophication of estuaries to acidification of forests, lakes, and streams. This Scope and Methods Plan focuses on five ecosystem effects on terrestrial and aquatic systems.

A.1.1 Terrestrial Nitrogen Enrichment

Deposited nitrogen compounds can act as a fertilizer to increase the productivity of plants and algae. However, too much nitrogen can lead to a surplus of nutrients, resulting in over-nutrient enrichment. **Table A-1** presents examples of nitrogen enrichment effects, which can impact species diversity by favoring nitrogen-tolerant species over other species that are more sensitive to nitrogen limits. Certain ecosystems may be dominated by species that have a competitive advantage in low nitrogen environments. When nitrogen increases, species that are normally kept in check by low nitrogen levels flourish and out-compete the other species in the community; thereby, potentially altering species' composition and diversity, nutrient cycling, and other ecosystem properties and functions. New plants may also move into ecosystems that are enriched in nitrogen, further challenging the native species. Animals that depend on specific plants for habitat and food may then be threatened by the changes to the plant communities that result from nitrogen inputs.

Table A-1. Examples of Nitrogen Enrichment Effects

Nitrogen Load (kg N ha-1• yr-1)	Nitrogen Enrichment Effects
>1.5	Altered algal communities in high-elevation freshwater lakes. (Colorado)
	Elevated N in tree leaf tissue high-elevation forests. (Colorado)
3 to 8	Mortality of sensitive lichen species. (West Coast)
5 to 35	A transect of 68 acid grasslands; species richness declines as a linear function of the rate of inorganic nitrogen deposition, with a reduction of one species per 4-m^2 quadrat for every 2.5kg N^{-1} yr^{-1}. (United Kingdom)
7	Community-level shifts in ombrotrophic bogs. (North Central and Eastern United States)
<8 to 10	N retained or denitrified (i.e., limited leaching). (many U.S. forests)
10 to 15	Change in plant species' competitive interactions lead to community-level shift in native grassland. (California)

Increased nitrogen deposition in western grass/shrub lands is also implicated in increased fire frequency in some areas because the nitrogen over-enrichment has favored the growth and production of fire-prone grass species. Emerging research has also linked increased nitrogen deposition to habitat alteration for threatened species.

Nitrogen enrichment, in combination with ozone exposure, causes major changes in tree health by reducing fine-root biomass and carbon allocation below ground and by greatly decreasing the lifespan of pine foliage. Nitrogen enrichment results in greater leaf growth, while ozone causes premature leaf loss at the end of the growing season. The net result of these pollutants is significant litter accumulation on the forest floor. Nitrogen cycling rates in soil are also stimulated by high nitrogen inputs, resulting in large leachate losses of nitrate from these watersheds and elevated fluxes of nitric oxide gas from soil. In coastal sage ecosystems that occur in low-elevation sites in this region, greenhouse and field studies indicate that nitrogen deposition may be one factor that enhances the invasion of exotic annual grasses.

A.1.2 Aquatic Nitrogen Enrichment and Eutrophication

Atmospheric deposition of nitrogen is a significant source of total nitrogen to many estuaries in the United States. National Oceanic and Atmospheric Association (NOAA) has calculated the amount of nitrogen entering estuaries ultimately attributable to atmospheric deposition for many of the East Coast estuaries. The amount of nitrogen entering estuaries due to atmospheric deposition varies widely, depending on the size and location of the estuarine watershed and other sources of nitrogen in the watershed. A number of uncertainties may result in a greater relative contribution of atmospheric deposition in some places. In addition, episodic inputs, which may be ecologically significant, may be higher than the annual average. Studies have shown that atmospherically deposited nitrogen (AD-N) contributes from 20 to >40% of the new nitrogen flux to estuaries and waters along the East Coast (Whitall and Paerl, 2001). The areas with the highest deposition rates stretch from Massachusetts to the Chesapeake Bay, and along the Central Gulf Coast.

The supply of nitrogen tends to limit the productivity of coastal ecosystems. However, nitrogen over-enrichment can alter a series of complex biogeochemical cycles that affect community processes (e.g., competition, community structure) and ecosystem processes (e.g., ecosystem efficiency, decomposition). Approximately 60% of estuaries in the United States

(65% of the estuarine surface area) suffer from over-enrichment of nitrogen, resulting in excessive algal growth, the outcome of which is eutrophication—the depletion of oxygen concentrations as the algae die and decompose. Symptoms of eutrophication include changes in the dominant species of phytoplankton (the primary food source for many kinds of marine life), low levels of oxygen in the water column, fish and shellfish kills, outbreaks of the toxic dinoflagellate, such as *Pfiesteria piscicida*, and cascading population changes up the food chain. In addition, encrustation and increased levels of turbidity in the water due to large amounts of algae can kill off submerged aquatic vegetation, which is an important habitat for many estuarine fish and shellfish species.

Often the most severe result of eutrophication is the depletion of dissolved oxygen (DO) in the water column by the decomposition of organic matter, produced within the ecosystem as a result of the nitrogen saturation. Anoxia (lack of oxygen) or hypoxia (DO concentrations lower than required by indigenous organisms) is a particular concern in coastal estuaries that exhibit density stratification (the division of water into layers with different temperatures and oxygen content), which occurs mostly during the summer months. Organic matter produced in lighted surface waters either sinks to the bottom waters where it decomposes, consuming oxygen inventories that are not replenished by photosynthesis, or it mixes with oxygen-rich surface waters. These low-oxygen zones may cause massive deaths of aquatic life and reduce the population densities of many important commercial fish and shellfish. Hypoxic bottom waters have expanded during the latter 20th century in many coastal ecosystems, including large areas of the continental shelf of the northern Gulf of Mexico near the mouth of the Mississippi River. Many of the highly eutrophic estuaries are along the gulf and mid-Atlantic coasts, overlapping many of the areas with the highest nitrogen deposition, but there are eutrophic estuaries in every region of the conterminous U.S. coastline.

Emerging ecological studies in the western United States demonstrate that some aquatic and terrestrial communities are significantly altered by increased nitrogen deposition. The major concerns are for ecological systems that are naturally adapted to low-nitrogen inputs, because increases related to anthropogenic atmospheric deposition can lead to nitrogen saturation. Where the inputs of nitrogen exceed the ecological system’s need for them, excess nitrogen is leached into surrounding waterways. Although much of the western United States is exposed to relatively low deposition of nitrogen, hotspots of nitrogen deposition occur downwind of large metropolitan areas or large agricultural operations. Some of the most sensitive systems affected by the elevated nitrogen deposition include high-elevation catchments in Colorado and chaparral catchments in the southwestern Sierra Nevada Mountains. The primary concerns in the West have been for the critical load of nitrogen deposition, which affects both terrestrial and aquatic resources through eutrophication and/or nitrogen enrichment, thereby altering community structure.

The over-enrichment of nitrogen deposition in high-elevation lakes has led to increased biomass of phytoplankton, resulting in eutrophication and shifts in diatom community composition. In the western United States, episodic acidification is also an important issue for surface waters throughout high-elevation areas. Where soils are sparse, as in alpine regions, most snowpack nitrogen is flushed to surface waters early in the snowmelt period. In addition to high-elevation lakes, snowpack nitrogen has also been reported to cause temporary acidification of alpine streams. Snowmelt-related temporary acidification of alpine lakes and streams and associated effects have been reported in numerous studies, largely for CAA Class 1 areas in the southern and central Rocky Mountains.

Increased nitrogen deposition in western arid and semiarid grass/shrub lands is also implicated in increased fire frequency in some areas because the nitrogen over-enrichment has favored the growth and production of fire-prone grass species. Emerging research has also linked increased nitrogen deposition to habitat alteration for threatened species. Nitrogen fertilization experiments in arid and semiarid plant communities have shown that changes in plant biomass associated with increased nitrogen deposition tend to alter species' composition, with negative impacts on biodiversity. Such plant community changes resulting from experimental fertilization have been reported in Joshua Tree National Park in California, coastal sage shrub communities of southern California, areas in the Chihuahuan Desert and Mojave Desert, and for arid grassland areas on the Colorado Plateau.

A.1.3 Aquatic Sulfur Enrichment

Atmospheric deposition of sulfur to primarily aquatic environments with high concentrations of organic matter leads to an increase in production of MeHg. The increased availability of sulfur as sulfate in lakes and streams due to the deposition of SO_x accelerates the conversion of elemental mercury and mercuric salts to highly toxic, bioaccumulative, and persistent MeHg compounds that build up in living tissue and increase in concentration up the food chain. Sulfate-dependant bacteria, along with methanogenic (methane-producing) microorganisms, are involved in the conversion of Hg^{2+} to MeHg under the anaerobic conditions found in wetlands, river sediments, and in certain soils. The presence of sulfates stimulates the growth of these methylating microbes. Acid rain is thought to increase biomethylation as more MeHg is formed under acidic conditions (less than a pH of 6).

The "cause-and-effect relationship" between sulfur and mercury deposition from the atmosphere has been demonstrated in the lab and in small-scale field experiments. Using sulfate levels equivalent to historical levels of sulfate deposition from acid rain in the northeastern United States, researchers in Minnesota have been able to the confirm a surge in MeHg levels with increasing levels of sulfates in a large-scale, fresh water wetland ecosystem (Jeremiason et al., 2006). While an in-depth assessment of mercury methylation relative to NO_x and SO_x inputs is not feasible at this time, reducing NO_x and SO_x inputs may also reduce the mercury methylation potential of some systems.

A.1.4 Terrestrial Acidification Due to Nitrogen and Sulfur

The current understanding of the effects of acidifying deposition on forest ecosystems has focused increasingly on the biogeochemical processes that affect plant uptake, retention, and cycling of nutrients within forested ecosystems. Research results from the 1990s indicate that decreases in base cations (e.g., calcium, magnesium, potassium) from soils are at least partially attributable to acid deposition in the northeastern and southeastern United States. Base cation depletion is a cause for concern because of the role these ions play in acid neutralization, as discussed above: In the case of calcium, magnesium, and potassium, these are essential nutrients for plant growth and physiology (e.g., nutrient uptake). As mobile aluminum increases due to soil acidification, the calcium/aluminum ratios change, which is partly related to the higher affinity of aluminum during passive uptake by roots. The change in these relative nutrient proportions has been correlated with declining forest health. Recent research indicates that the loss of cations also leads to aluminum leaching from the soil to stream waters, which can have harmful effects on fish.

The loss of calcium from forest soils and forested watersheds has now been documented as a sensitive, early indicator of the soil response to acidifying deposition for a wide range of forest soils in the northeastern United States (Likens, Driscoll, and Buso, 1996). There is a strong relationship between acid deposition and leaching of base cations from soils in hardwood forests (e.g., maple, oak), as indicated by long-term data on watershed mass balances, plot- and watershed-scale acidification experiments in the Adirondack Mountains and in Maine, and studies of soil solution chemistry along an acid-deposition gradient from Minnesota to Ohio.

A.1.5 Aquatic Acidification Due to Nitrogen and Sulfur

Acidifying deposition causes acidification of sensitive surface waters. The effect of acidifying deposition on aquatic systems depends largely upon the ability of the ecosystem to neutralize the additional acid. This is referred to as an ecosystem’s ANC.

ANC levels depend largely on a watershed’s physical characteristics: geology, soils, and size. Water systems that are sensitive to acidification tend to be located in small watersheds that have few alkaline minerals and shallow soils. Large, forested watersheds have been shown to acidify during large rainfall and snowmelt episodes. As acidity increases, aluminum leached from soils and sediments flows into lakes and streams and can be toxic to aquatic species. The lower pH levels and higher aluminum levels that result from acidification make it difficult for some fish and other aquatic species to survive, grow, and reproduce. In some waters, the number of fish species able to survive has been directly correlated to water acidity. Acidification can also decrease fish population density and individual fish size.

In western regions, some high-elevation lakes, particularly in the Rocky Mountains, have become acidic, especially during snowmelt. However, while many western lakes and streams are sensitive to acidification, they are not subject to continuously high levels of acid deposition, and, therefore, have not become chronically acidified. During the 1980s and 1990s, an integrated study of atmospheric deposition, terrestrial ecosystems, and aquatic ecosystems was conducted in several watersheds in the Sierra Nevada Mountains to determine if acidifying deposition was affecting these areas and to infer the implications of acidification on surface waters in the region. Chronic acidification of high-elevation surface waters in the Sierra Nevada Mountains was not found, but episodic changes in stream water chemistry did occur. In many of the watersheds studied, for example, the pH decreased as the waters became more acidic with increasing runoff, reaching a minimum during peak snowmelt.

Anthropogenic atmospheric nitrogen and sulfur deposition impacts the surface seawater chemistry by increasing acidification and reducing alkalinity. The impacts of these changes are more substantial in coastal waters where the ecosystems are more vulnerable due to other human impacts, such as nutrient enrichment and pollution. Ocean acidification is a significant threat to coral reefs and coastal benthic and planktonic foodwebs which, in turn, impacts fish populations.

A.2 Additional Effects Related to Acidification and/or Nutrient Enrichment on Sensitive Ecosystems

It important to note that several additional indicators are worthy of investigation:

- Nitrogen saturation
- Maple decline
- Ammonia air deposition and toxicity to native mussels
- Relationship between acidity/nutrient enrichment and mercury methylation
- Sensitive areas for acidity/nutrient enrichment impacts.

A concise summary of information on each topic is introduced in the following sections.

A.2.1 Nitrogen Saturation

Terrestrial and aquatic ecosystems in the eastern and northeastern United States receive nitrogen deposition loadings related to air pollution that are far in excess of natural background levels. When combined with other acid rain impacts to soil systems and receiving waters, the terrestrial ecosystem, in particular, can become nitrogen saturated. This leads to the increased "leakage" of nitrogen into groundwater and surface water. Studies are now documenting similar concerns for sensitive areas that often involve national parks in the Appalachians (e.g., Smoky Mountains National Park), the Rocky Mountains, the Cascades, and other areas in the West. Several forests throughout the United States are beginning to show signs of nitrogen saturation, a condition where the inputs of nitrogen exceed the forest's need for them and excess nitrogen is leached into surrounding waterways. Significant studies include those identified by Driscoll and colleagues (2001); Fenn and colleagues (2003); and NAPAP (2005).

A.2.2 Maple Decline

Maple decline is a generalized term for a set of symptoms that may be applied to any species of tree suffering a wide range of different stressors, resulting in a loss of vigor or habitat.

These symptoms have been studied for the northeastern United States in recent decades, with a focus on native maples, such as the sugar maple. In rural areas, maple decline is often attributed to soil acidification caused by acid rain. Soils that have developed from nutrient-poor parent materials, such as sandstone, quartzite, and granite are most sensitive to acidification. Some of the best-documented work has been conducted at the Hubbard Brook Experimental Forest (HBEF), a 3,160-hectare reserve near North Woodstock, NH, where scientists have measured soil composition for the past 50 years. A recent study (Juice et al., 2006) documents how scientists added nutrients in a test plot to replicate soil conditions that existed prior to the loss of sugar maples over the past 25 years. This reproduced the favorable soil conditions that existed prior to 20th-century industrial pollution, with the result that sugar maples on the test plot rebounded dramatically. Nitric and sulphuric acid in acid rain leaches calcium from the soil. Calcium is the second most abundant plant nutrient after nitrogen. In addition, the loss of calcium leads directly to acidic soils. When soils become too acidic, trees such as sugar maples become stressed and have a harder time growing or producing seeds and seedlings.

A.2.3 Ammonia Air Deposition and Toxicity to Native Mussels

Across North America, populations of freshwater mussels have fallen drastically to the point where more than 70% of native unionid mussel species are considered endangered, threatened, or of special concern. Ammonia toxicity is of concern for the survival of juvenile mussels (the most sensitive life stage), and EPA’s Office of Water (OW) is pursuing studies to consider revisions to its total ammonia criterion guidance to provide protections against acute or chronic toxicity (U.S. EPA, 2005).

A.2.4 Relationship between Acidity/Nutrient Enrichment and Mercury Methylation

Research recently summarized for the northeastern United States suggests a relationship between sulfur deposition and processes promoting mercury methylation, which increases the risks for bioaccumulation and biomagnification in food chains. Ongoing research in the Gulf of Mexico is also addressing the following three research goals:

- Research Goal 1 – Test the hypothesis that rates of MeHg production in coastal sediments are in part controlled by temporal and spatial hypoxia patterns that result from coastal eutrophication, and that maximum MeHg production occurs in regions adjacent to hypoxic zones.

- Research Goal 2 – Test the hypothesis that coastal eutrophication and hypoxia can result in elevated MeHg accumulation and biomagnification in red snapper and gray snapper, both commercially and recreationally important fish species in this region.
- Research Goal 3 – Test the hypothesis that anglers who proportionately consume fish from areas of higher MeHg production related to hypoxia will have higher rates of mercury exposure (as measured by the concentration of mercury in hair) than anglers consuming similar amounts of fish from other coastal Louisiana locations where hypoxia does not occur.

The EPA ORD has also done some preliminary work on developing large-basin multimedia modeling systems that would handle impacts from nutrients (especially nitrogen species) and the complicated processes related to mercury. Significant sources identified include Chesney and colleagues (2006), Driscoll and colleagues (2007), and Evers and colleagues (2007).

A.3 Sensitive Areas for Acidity/Nutrient Enrichment Impacts

Research accomplished through the NAPAP (2005) has identified spatial areas in terms of both terrestrial and aquatic ecosystems that show sensitivity to impacts from acid rain–related air pollution (**Figure A-1**). The NAPAP has been able to develop GIS layers of such sensitive areas as ecoregions or hydrologic basins.

The degree to which other types of sensitive areas (related to such themes as nitrogen saturation, maple decline, or the locations of waterbodies showing nitrogen limitations) can be at least robustly defined in terms of their geographical extents could be examined. Where such geospatial data stratification materials can be identified, this would be helpful in both the analysis of indicators and in the development and interpretation of results from models. An examination of these materials would also be useful in identifying critical loads for acidification and nutrient enrichment stressors, which in most cases will vary for different ecoregions and waterbody types.

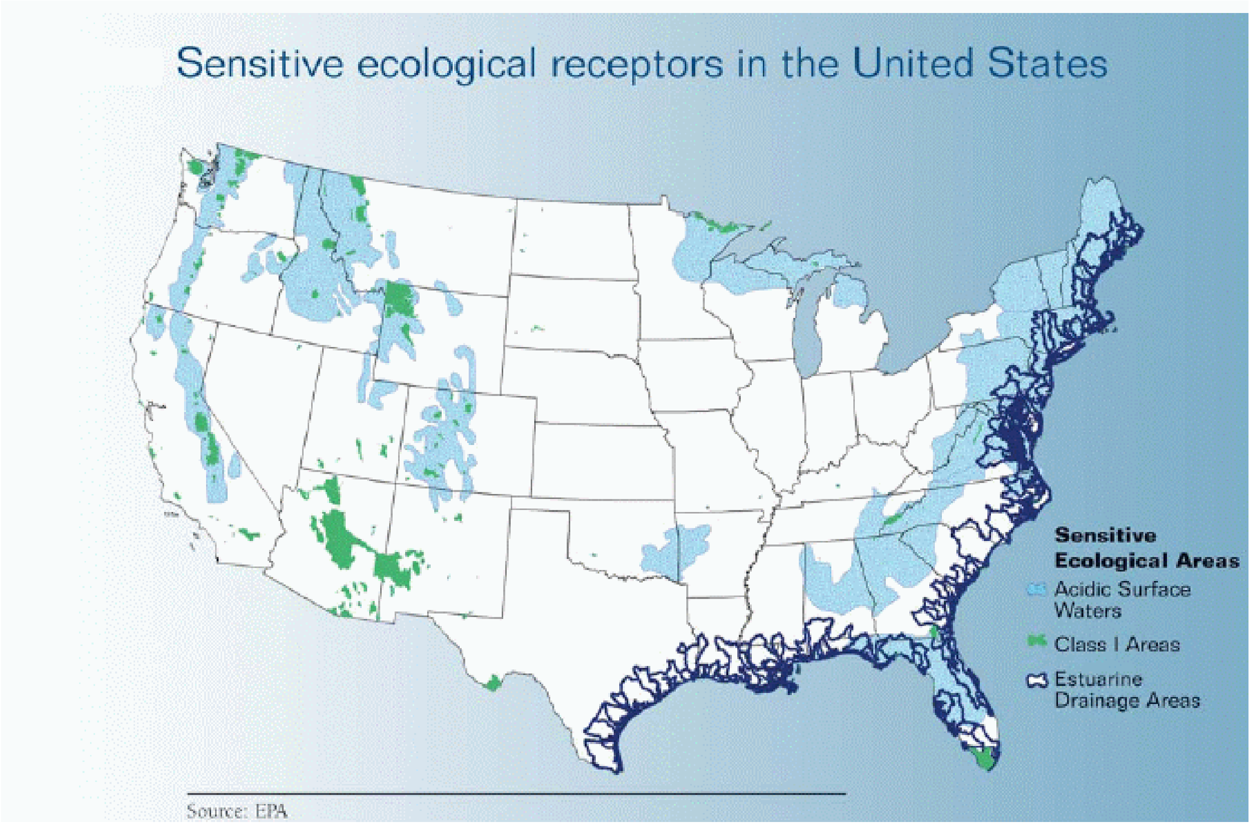

Figure A-1. Sensitive ecological receptors in the United States.

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APPENDIX B

OTHER INDICATORS – NITROUS OXIDE

NAAQS' purpose to protect welfare entails not only ecological welfare, but also other forms of welfare that are potentially impacted, such as materials, climate change, and visibility. N_2O is a naturally occurring greenhouse gas; however, human activities have increased atmospheric concentrations by 18% since the preindustrial era (IPCC, 2001). The global warming potential of CO_2 is 1, while the global warming potential of N_2O is 310, indicating that a molecule of N_2O is 310 times more effective in trapping heat in the atmosphere than CO_2 over a 100-year period. Based on the current U.S. Greenhouse Gas (GHG) Inventory (U.S. EPA, 2007), nitrous oxide contributes approximately 6.5% to total greenhouse gas emissions (in CO_2 equivalents)

Elevated nitrogen loading to ecosystems can significantly enhance the production of N_2O. For example, the nitrogen nutrient loading in water bodies can enhance N_2O emissions from the bacterial breakdown of nitrogen from these sources. In another example, numerous studies have shown that N_2O emissions from soils increase upon artificial nitrogen additions (Brumme and Beese, 1992; Matson et al., 1992; Klemedtsson et al., 1997; Papen et al., 2001). Regions with elevated atmospheric nitrogen deposition due to anthropogenic activity also show increased N_2O emissions (Butterbach-Bahl et al., 1998, 2002). Nitrous oxide emissions from soils are also influenced by precipitation and temperature. The Photosynthesis-Evapotranspiration-Model–Nitrogen–Denitrification-Decomposition (PnET-N-DNDC) model is designed to simulate and predict soil carbon and nitrogen biogeochemistry in temperate forest ecosystems and to simulate the emissions of N_2O and NO from forest soils. The model couples the PnET model, the DNDC model, and a nitrogen module that are further described in Li and colleagues (1992, 1996, 2000), Li (2000), and Stange and colleagues (2000). The capacity of this model to simulate nitrogen trace gas emissions from forest soils was tested by comparing model results with results from field measurements at 19 different field sites across Europe and 1 site in the United States (Kesik et al., 2005). Denitrification is described in the model as a series of sequential reductions driven by a microorganism using nitrogen oxides as electron acceptors under anaerobic conditions. As intermediates of the processes, NO and N_2O are tightly controlled by the kinetics of each step in the sequential reactions. Interactions between temperature, precipitation, and forest soil NO and

N_2O emissions in Europe were investigated using PnET-N-DNDC (Kesik et al., 2006) abiotic parameters that are included in the model and are summarized in **Table B-1**.

Table B-1. Parameters Included in the PnET-N-DNDC Model as Modeled for European Forest (Kesik et al., 2006)

Forest Properties	Soil Properties	Daily Climate Input Parameters	Tree Species/Genera
Forest type Age Above and below ground biomass Plant physiology parameters	Texture Clay content pH Soil organic carbon content Stone content Humus type	Precipitation Min and max air temps Inorganic [N] in Precip	Pine Spruce Hemlock Fir Oak Birch Beech Slash Pine Larch Cypress Evergreen Oak

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APPENDIX C

CMAQ MODELING

The 1998 *Guidelines for Ecological Risk Assessment* (U.S. EPA, 1998) state that "At the beginning of the analysis phase, the [risk] assessor critically examines the data and models to ensure that they can be used to evaluate the conceptual model developed in problem formulation." The assessor evaluates the strengths and limitations of different types of data and evaluated measurement or modeling studies to be used, including accounting for uncertainty. This section presents our current thoughts for using existing ambient air quality and deposition monitoring data, as well as the application of atmospheric and multimedia models to predict ecological risk and exposure.

C.1 Atmospheric Modeling

The atmospheric processes that transform and transport ambient NO_x and SO_x to deposition species are similar. (NOTE: In addition, NO_x and SO_x interact photochemically in the PM formation. Further, since NO_x participates in both the ozone and PM photochemistry, those pollutants are linked and need to be simulated together in a single model.) Therefore, we intend to use the same process model for atmospheric fate, transport, and deposition. We plan to use the CMAQ model, a peer-reviewed, state-of-the-art model of the atmosphere. The 2002-based CMAQ modeling platform will likely be used as the tool for the air quality modeling.

As shown in **Figure C-1,** the CMAQ modeling domain covers the continental United States and portions of Canada and Mexico. There are two 12 x 12 km horizontal-grid resolution modeling domains, an eastern United States domain (outlined in red), and a western United States domain (outlined in blue). The modeling domain contains 14 vertical layers, with the top of the modeling domain at about 16,200 meters.

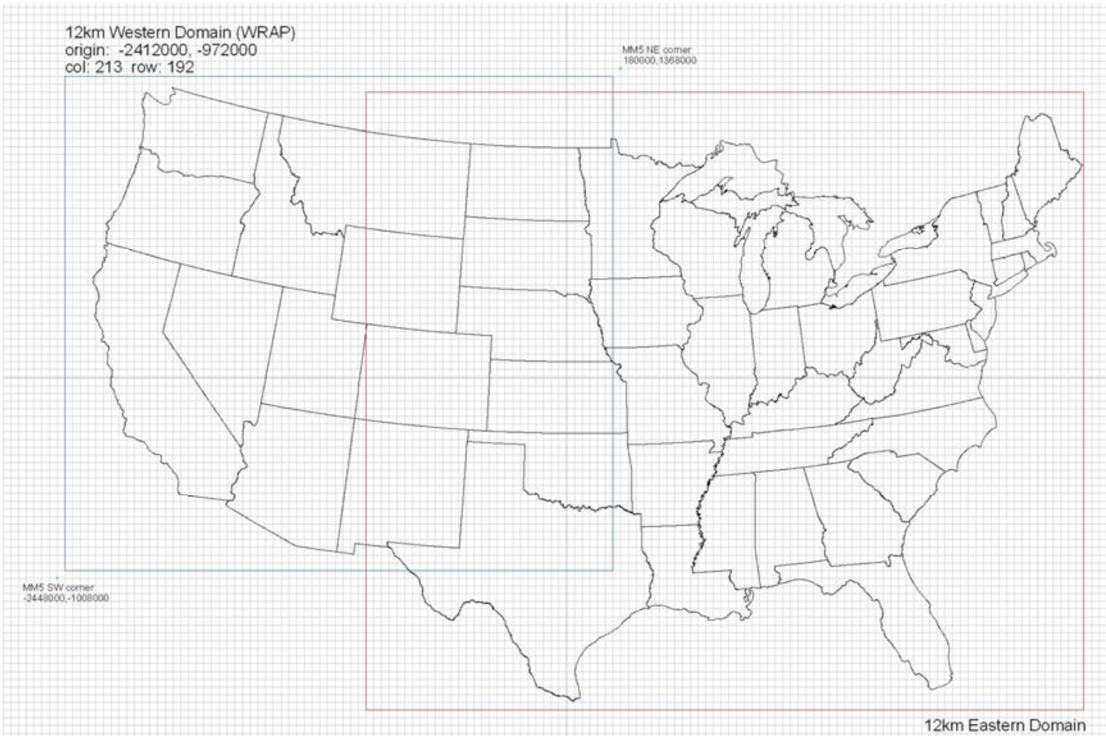

Figure C-1. CMAQ 12-km eastern and western United States modeling domains.

The key inputs to the CMAQ model include emissions from anthropogenic and biogenic sources, meteorological data, and initial and boundary conditions. The CMAQ meteorological input files were derived for the entire year of 2002 from a simulation of the Pennsylvania State University/National Center for Atmospheric Research Mesoscale Model (Grell et al., 1994). This model, commonly referred to as MM5, is a limited-area, nonhydrostatic, terrain-following system that solves for the full set of physical and thermodynamic equations, which govern atmospheric motions. Anthropogenic and biogenic emissions from the 2002 CMAQ modeling platform were obtained from the 2002 National Emissions Inventory (NEI). These data were then processed using the Sparse Matrix Operator Kernel Emissions (SMOKE) processing system.

In addition to understanding the relationship between ambient concentrations and deposition, we also need to understand how the existing patterns of deposition modeled using CMAQ may vary with variability in meteorology (e.g., temperature, precipitation). By making use of the 5 years of CMAQ simulations conducted for the joint EPA/Centers for Disease Control and Prevention (CDC) project, we may be able to provide a quantitative assessment of the variability in deposition patterns induced by the meteorological variability between years. This assessment plans to include statistical analyses designed to compare the magnitude and spatial patterns of deposition for sulfur and nitrogen containing species predicted by CMAQ across the 5 years modeled. The analysis intends to highlight the similarities and differences in the distribution of monthly, seasonal, and annual wet, dry, and total deposition for grid cells covering selected watersheds, forested ecosystems, and croplands. Specifically, for each geographic area we plan to calculate (1) the 5-year average in monthly, seasonal, and annual nitrogen and sulfur deposition and (2) the range in the deposition for each of these time periods within the 5 years modeled. This information may be presented in tabular and graphical forms to reveal (1) how deposition varies by year, season, and month for the 5 years, on average, and (2) how monthly and seasonal deposition in the individual years compares to the 5-year average. We may also examine the spatial variation in deposition within and between the selected geographic areas. For this we plan to prepare histograms showing the distribution of deposition by month and season across the multiple grid cells within each of the selected geographic areas. The results may be analyzed to compare the distribution and range of values for the different areas. The feasibility of using statistical techniques to quantify year-to-year variability in the spatial patterns of predicted deposition within a geographic area may be explored.

C.2 Ambient Air Concentration Data and Deposition Data

Because the risk/exposure assessment will ultimately be used to assess current and alternative ambient NO_x and SO_x standards, the risk assessment will need to characterize both the effects associated with the deposition of nitrogen and sulfur and the relationship between ambient concentrations at a specific geographic resolution and deposition at "downstream" receptors. **Figure C-2** shows the flow of information and modeling we propose to use to characterize the ambient levels of NO_x and SO_x and estimates of nitrogen and sulfur deposition.

As shown in **Figure C-2**, in addition to CMAQ modeling, we plan to evaluate the available observational data on deposition for use in characterizing ambient and deposition surfaces across the United States. There are a number of databases of ambient monitoring data, most of which are collected in EPA's Air Quality System (AQS). There are also databases of deposition monitoring data, including the NADP, the CASTNet, and the Park Research and Intensive Monitoring of Ecosystems NETwork (PRIMENet).

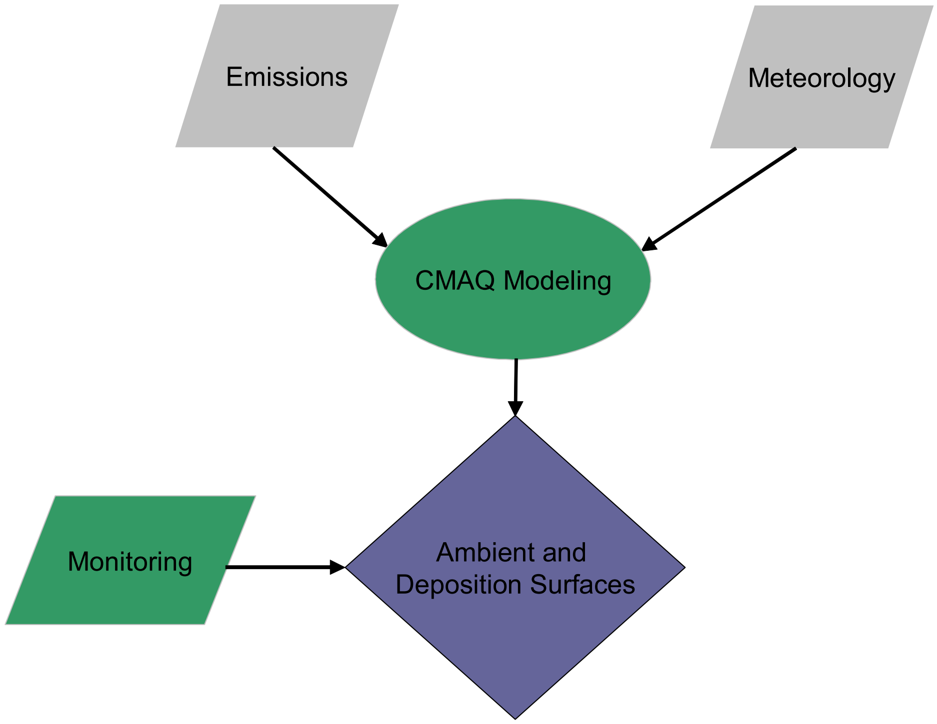

Figure C-2. Development of ambient NO_x and SO_x and nitrogen and sulfur deposition characterizations.

We plan to assess the feasibility of combining data from modeling and monitoring to produce ambient air quality and deposition surfaces that are grounded in observational data but make use of the models' ability to capture the impacts of emissions and meteorology on the geographic gradients of air quality and deposition. To assess the feasibility of combining data, we intend to implement one or more methods on data associated with a region of interest to this project. The estimated air quality and deposition surfaces will be helpful in assessing baseline risks and serve as baselines for assessing the impacts of attaining alternative standards.

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APPENDIX D

MULTIMEDIA MODELING

Atmospheric modeling for this quantitative risk/exposure assessment intends to use, where possible, the CMAQ modeling system. The outputs from the CMAQ model must be linked to one or more multimedia models (e.g., models for terrestrial systems or aquatic systems) to determine the environmental effects of atmospheric deposition of NO_x and SO_x. For this analysis, there is a need to consider models that can potentially handle all types of total reactive nitrogen (e.g., consideration of such species as ammonia and nitrous oxide) and models that can combine analyses of sulfur with mercury methylation processes.

The choice of multimedia models to determine and quantify these effects is not as clear as in the atmospheric modeling. With multimedia models, there are numerous technical formulations, parameterizations, and geographic considerations to consider in choosing the correct model for the task at hand. This decision depends on desired outcomes and simulated parameters. Outcomes may range from quantified, speciated sulfur or nitrogen concentrations in the soil and surface water to measures of species' richness in a forest landscape. Not all effects can be estimated with multimedia models, so a decision on the models to use must balance the pros and cons of the models that are available.

D.1 Model Characterization

The 35 models listed in **Table D-1** represent a wide diversity of types of ecosystems; history, location, and spatial/temporal scale of application; scientific acceptance and organizational and agency support; complexity and requirements; state variables and processes; and management uses. Each model was examined to elucidate the necessary information for choosing the appropriate multimedia model. Of the 35 models, 9 are classified as receiving-water models, and 26 are classified as terrestrial/watershed models, although these distinctions are somewhat arbitrary. In recognition of some models having enough common features or history of integration to warrant merging of the discussion, the table also presented several models together; however, although presented collectively, these models may not share the same history, theory, development, and application.

Table D-1. Information Detailed for Each Model of Interest

Model Information	
Name	Model type
Description	Indicators
Supporting organization(s)/agency endorsements	Resource requirements
Scientific acceptance	Data requirements
Type of ecosystems modeled	Model performance/evaluation
Where has it been applied?	Model technological integration
Spatial scale	Key environmental/ecological processes
Transferability	Management use
Temporal features	Citations/URLs
Model inputs related to deposition	Notes (miscellaneous)

Table D-2 presents a more concise overview of the models identified; information is presented that summarizes both the type of ecosystem that the model has been designed to represent and the relative level of complexity with which the model considers system components and processes. Model complexity was measured by the underlying theory and the effort necessary to parameterize a model. For instance, an empirical model that requires only basic nutrient information and relies on many assumptions would constitute a low level of complexity, whereas a mechanistic model with a large number of variables to parameterize would constitute a highly complex model. Complexity does not necessarily suggest desirability, utility, or credibility, as these concepts are highly context-specific. All of the models presented have arguably demonstrated substantial technical acceptance based on their historical use, presence in peer-reviewed publications, and degree of agency support. Further discernment regarding “quality” or applicability of the different models to support elements of the secondary NO_2 and SO_2 NAAQS should carefully consider a variety of contextual drivers when using an integrated modeling approach.

Table D-2. Supported Model Components And Level Of Representation

	Model	Urban	Rural	Agriculture	Forest	River	Lake	Reservoir	Estuary	Coastal	Groundwater
WATER MODELS	AQUATOX	—	—	—	—	■	■	■	—	—	—
	QUAL2K	—	—	—	—	◙	—	—	—	—	—
	WASP7	—	—	—	—	■	■	■	■	■	—
	CE-MODELS	—	—	—	—	■	■	■	■	■	—
	ILWAS	—	□	—	◙	◙	◙	◙	—	—	□
	LWWM	■	—	—	—	■	■	■	■	■	◙
	RCA	—	—	—	—	■	■	■	■	■	—
	THMB/IBIS	—	◙	◙	◙	◙	—	—	—	—	◙
	WARMS	—	—	—	—	—	◙	◙	—	—	—
WATERSHED/TERRESTRIAL MODELS	HSPF/LSPC	■	■	■	■	□	□	□	—	—	□
	PLOAD	□	□	□	□	—	—	—	—	—	—
	SWAT	◙	■	■	■	■	◙	◙	□	—	■
	WARMF	■	■	◙	■	■	■	■	—	—	◙
	BIOME-BGC	—	■	◙	■	—	—	—	—	—	—
	PnET-BGC	—	◙	—	◙	◙	◙	◙	—	—	—
	CENTURY 4/ CENTURY 5	—	■	■	■	—	—	—	—	—	□
	DayCent	—	■	■	■	—	—	—	—	—	□
	DNDC	—	□	◙	□	—	—	—	—	—	—
	DRAINMOD	—	□	■	—	□	—	—	—	—	◙
	EPIC/APEX	—	◙	■	□	—	—	—	—	—	□
	GLEAMS	—	◙	■	□	—	—	—	—	—	◙
	Hole-in-the-pipe	—	■	—	■	—	—	—	—	—	—
	INCA	—	◙	◙	◙	□	—	—	—	—	—
	MAGIC	—	◙	—	◙	◙	◙	◙	—	—	◙
	MERLIN	—	■	□	■	—	—	—	—	—	□
	NLOAD/NLM/ELM	□	□	□	□	—	—	—	□	—	□
	NuCM	—	◙	—	■	—	—	—	—	—	—
	ReNuMa	◙	◙	◙	◙	—	—	—	—	—	□
	NWPCAM	□	□	□	□	□	—	—	—	—	—
	RHESSys	—	■	■	■	□	—	—	—	—	—
	SAFE	—	■	—	■	—	—	—	—	—	◙
	Simple Mass Balance	□	□	□	□	□	□	□	□	□	—
	SPARROW	□	□	□	□	□	—	□	—	—	—
	TOPMODEL or GT/MEL	—	◙	◙	◙	◙	—	—	—	—	—
	WATERSN	□	□	□	□	—	—	—	—	—	—

— Not supported
□ Relatively simple representation of features and processes
◙ Moderate level of representation of features and processes
■ Most complex representation of features and processes

D.2 Multimedia Model Selection

There are four basic steps necessary to undertake a modeling effort to examine the effects of nitrogen and sulfur deposition (RTI 2007):

1. Choose the specific question/problem to address.
2. Choose the best models based on model formulation (e.g., are biological processes considered?), desired output, study area, data availability, and necessary uncertainty/sensitivity analyses for the models.
3. Determine and set up any processes/algorithms necessary to match atmospheric modeling output (assumed to be from CMAQ) to the chosen receiving water or terrestrial/watershed model.
4. Obtain the data needed for model parameterization.

The model details presented in the summary table and concluding discussion should assist the reader in making an informed decision on the best model for a task. The difficulty with this area of work lies with the desire to utilize atmospheric modeling in combination with the receiving-water and terrestrial/watershed models. The multi-media approach to modeling is still in development, so, at this time, not many models are set up to immediately accept the output from an atmospheric model such as CMAQ. Several of the models examined accept atmospheric concentration or flux data, but the time-step, spatial resolution, and exact species required might all differ from the atmospheric model output. For those models that accept atmospheric inputs in a form other than that output by CMAQ, efforts can be made to reconcile the outputs of the atmospheric model and the inputs of the ecosystem model.

Most applications for the determination of acidification effects have taken place in the northeastern United States because of the interest in quantification of acid rain effects. A specific effort was made to determine which models of the 35 could apply to other regions of the United States.

Review of past applications and technical documentation for each of the 9 receiving water models reveals that 4 of the 9 models are accepting of parameters for all regions of the country. These models are: AQUATOX, the CE-QUAL family of models, QUAL2K, and WASP. The WARMS model has been used in Canada; therefore, it may be applicable in more alpine regions of the United States. The remaining 4 models have either been developed for a specific area of the country or show limitations in their extension beyond the regions in which they have already been applied.

A listing of the terrestrial/watershed models that show promise in applications across the United States, specifically in western states, follows with a short explanation of support.

- BIOME-BGC – validated with western locations, including arid, cold, western climates
- CENTURY 4/5 – has been used in the Great Plains and Rocky Mountain areas of the United States
- DayCent (DayCent-Chem) – validated in an alpine/subalpine watershed of Colorado and used at other sites across the United States
- EPIC/APEX – used across the country, but only for agricultural lands
- GT/MEL – a relatively new model that has been tested in the western Oregon Cascades
- HSPF – used in applications across the country
- MAGIC – used worldwide and on many types of systems but does not include biological processes directly and lacks features to simulate processes involving nitrogen
- Mass balance – may be applied anywhere data are available
- MERLIN – no applications uncovered for U.S. sites but has been used in locations in Europe
- PLOAD – a screening-level model that may be applied anywhere data are available
- PnET-BGC – many applications in the northeastern United States; developed for forest landscapes
- RHESSys – used in applications in the mountains of Montana
- SAFE – wide applications in forest systems
- SWAT – used throughout the country, but does not readily accept the atmospheric deposition inputs required for this study, although a workaround may be possible
- WARMF – used across the country (currently utilizes atmospheric concentration data; a newly undertaken project will modify WARMF to accept atmospheric deposition fluxes to make it more compatible with CMAQ).

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APPENDIX E

VALUATION OF ECOSYSTEM SERVICES

E.1 Introduction to Ecosystem Services

The definition of adverse is “unfavorable or antagonistic in purpose or effect.” One way to assess adverse effects on welfare is through quantification of ecosystem services. The adverse effect would be the loss or reduction of those services through the effects of NO_x and SO_x on the underlying ecological processes and functions that constitute the service. EPA defines ecosystem services as the outputs of healthy, intact ecosystems and the underlying ecosystem processes and functions that contribute to human well-being (U.S. EPA, 2006). As articulated by the Millennium Ecosystem Assessment (United Nations, 2005) from the United Nations, these include provisioning services (e.g., clean water, food, wood, fiber, fuel), regulating services (e.g., water purification, climate regulation), supporting services (e.g., nutrient cycling, soil formation), and cultural services (e.g., recreation, spiritual). Regulating services are of key importance to EPA because they directly impact air and water quality, and they have strong links to human heath and well-being. Therefore, assessing changes in ecosystem services may be one means of assessing whether an effect is adverse. Key issues are how to aggregate across different ecosystem services or how to select a representative ecosystem service that is most sensitive to deposition effects. Some potential indicators of ecosystem services include the quality of a critical habitat, biodiversity, species composition, controlling/limiting invasive species, and pest outbreaks. Determining the exposure-response relationships of NO_x and SO_x on the ecosystem service and the underlying ecological process and function will provide a broader focus in determination of adverse impacts. For more background information on ecosystem services, see *The Integrated Review Plan for the Secondary National Ambient Air Quality Standards for Nitrogen Dioxide and Sulfur Dioxide* (U.S. EPA, 2007).

Economic Valuation. A succinct statement of the economic approach to valuation is in the Ecological Benefits Assessment Strategic Plan (EBASP) (U.S. EPA, 2006): “Economists generally attempt to estimate the value of ecological goods and services based on what people are willing to pay (WTP) to increase ecological services or by what people are willing to accept (WTA) in compensation for reductions in them. To enable a comparison of policy options, a common unit is needed to express the value of ecological goods and services. The dollar is the preferred unit for valuation, because there is an extensive body of literature addressing its application and interpretation and it is easily compared with costs for considering the net effects of alternative policy choices. Three primary approaches for estimating these values exist: market-based methods, revealed preference methods, and stated preference methods (U.S. EPA, 2006)." The EBASP document continues by further explaining these types of economic monetary valuation methods.

For market-based valuation, for many regions of the country, there are diverse crop assemblages, including timber for building materials, timber for pulp and paper, biofuel crops, grain and soybean feed crops, grass seed, orchard crops, row crops, Christmas trees, and horticultural crops. There are numerous methods and data for valuation of these market commodities.

A useful approach to pursue with economists would be mapping the monetary values of ecosystem services by land use across the ecosystem service district, using the approach described in Troy and Wilson (2006). This approach will first require mapping biophysical measurements of services as discussed in Section 3.2. Then, economics expertise will be needed to apply monetary values to these services as well as to other services, for which there are transferable methods and values from studies elsewhere, using the "Environmental Valuation Reference Inventory" ([http://www.evri.ca/english/default.htm] and other sources of information).

Biophysical Valuation. Because monetization of many ecosystems services is either very difficult or problematic for a number of reasons, nonmonetary valuation using biophysical measurements and concepts will be pursued in addition to economic valuation. One approach that fits well with the empirical and modeling data is what has been called relative value indicators or relative benefit indicators (Wainger et al., 2001; Boyd and Wainger 2003). In this approach, a set of indicators is defined that reflect site and landscape features that affect (1) the quality and quantity of ecosystem functions or services, (2) the availability of complementary goods and services, (3) the scarcity of goods and services, and (4) the reliability of the flows of ecosystem services. These indicators are then aggregated into a combined site or place-based index using standardized scoring (Wainger et al., 2001). This is an explicit approach to a “bundling” of services. The development of ERFs and ETFs in Section 3.7 may be adapted to such an approach.

A related approach derives from an economic method that develops “productivity indexes” and now calls these “environmental performance indexes” (Färe et al., 2004). This approach attempts to combine positive and negative outcomes from a system (e.g., factory, region, county) into a single index of performance. To the degree that this approach is science-based, it can be developed with available data and offers potential in a qualitative risk assessment.

A second approach to biophysical valuation assigns values to ecosystem goods and services through the use of the common currency of energy. This approach has been supported in part by the EBASP (U.S. EPA, 2006) and other reviews of valuation. A comprehensive, holistic method of energy-based valuation is the energy systems analysis of environmental accounting developed by H. T. Odum (1996). A similar approach has been developed by Bakshi and colleagues (Hau and Bakshi, 2004a, 2004b; Ukidwe and Bakshi, 2004, 2005). There is extensive literature supporting and elaborating on these approaches.

By describing the differing bundles of ecosystem services under varying levels of NO_x and $SO_{x,}$ and offering choices between those bundles, tradeoff or indifference curves can also be generated reflecting individual- and population-level preferences for different ecosystem functions. These tradeoffs can be presented to survey respondents using methods such as conjoint analysis to provide measures of preferences for different levels of ecosystem functions as expressed through ecosystem service levels**. Table E-1** shows an example of how ecosystem services information might be presented to a survey respondent in a conjoint analysis framework.

The results of a conjoint analysis can be used to convert all ecosystem services to a common unit based on preference weightings for different types of services. This may allow ecosystem services to be combined for comparing impacts across alternative forms or levels of NO_x and SO_x standards.

Table E-1. Example Survey Design for Preference-Based Ecosystem Tradeoffs

Example Conjoint Scenario Rating Form

Ecosystem Services Scenario Descriptions				
	Water Quality	Habitat	Food and Fiber Production	Carbon Sequestration
Scenario 1	Drinkable	Low diversity	High	High
Scenario 2	Drinkable	High diversity	Low	Low
Scenario 3	Swimmable	Medium diversity	Medium	Medium
Scenario 4	Boatable	Low diversity	High	Medium
Scenario 5	Swimmable	Low diversity	Low	Low

Scenario Rating
Please rate how desirable each scenario is overall by circling one of the numbers in each row of the following table:

	Highly Desirable	Quite Desirable	Desirable	Slightly Desirable	Neither Desirable nor Undesirable	Slightly Undesirable	Undesirable	Quite Undesirable	Highly Undesirable
Scenario 1	1	2	3	4	5	6	7	8	9
Scenario 2	1	2	3	4	5	6	7	8	9
Scenario 3	1	2	3	4	5	6	7	8	9
Scenario 4	1	2	3	4	5	6	7	8	9
Scenario 5	1	2	3	4	5	6	7	8	9

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United States Environmental Protection Agency	Office of Air Quality Planning and Standards Health and Environmental Impacts Division Research Triangle Park, NC	Publication No. EPA-452/D-08-002 March 2008