UNITED STATES ENVIRONMENTAL PROTECTION AGENCY
WASHINGTON D.C. 20460

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SCIENCE ADVISORY BOARD

December 23, 2008

EPA-CASAC-09-004

The Honorable Stephen L. Johnson
Administrator
U.S. Environmental Protection Agency
1200 Pennsylvania Avenue, N.W.
Washington, DC 20460

Subject: Peer Review of EPA's *Risk and Exposure Assessment to Support the Review of the Secondary National Ambient Air Quality Standard for Oxides of Nitrogen and Sulfur: First Draft*

Dear Administrator Johnson:

The Clean Air Scientific Advisory Committee (CASAC) NO_x & SO_x Secondary National Ambient Air Quality Standards (NAAQS) Review Panel held a public meeting on October 1-2, 2008 to review EPA's *Risk and Exposure Assessment to Support the Review of the Secondary National Ambient Air Quality Standard for Oxides of Nitrogen and Sulfur: First Draft* (see Enclosure A for the Panel roster). The Panel discussed its draft report on November 19, 2008 at a public teleconference. The Panel's draft report was reviewed and approved by the chartered CASAC at the public teleconference on December 19, 2008 (see Enclosure B for the CASAC roster).

Overall, the Panel found the first draft of the Risk and Exposure Assessment (REA) document to be a credible beginning in the development of the assessments needed to support future rule-making. The Panel recognizes the time constraints under which the present first draft REA was prepared and that much of the analyses and the subsequent interpretation have yet to be completed (e.g., Chapter 7 is focused primarily on acidifying deposition and Chapter 8 only begins to explore how a secondary NAAQS might be structured). The Panel has many suggestions for strengthening the document in response to the Agency's charge questions. Individual comments from the Panel members are provided in Enclosure C.

The Panel strongly supports the up-front inclusion of a policy-focused interpretation of each of the key scientific findings in the Integrated Science Assessment (ISA). These policy interpretations should be included in an Executive Summary, as well as in the applicable sections of the REA. As discussed in prior reviews of the ISA and Scope and Methods documents, it is critical that the REA give attention to all forms of reactive nitrogen (Nr, which includes oxidized, chemically-reduced and organic nitrogen forms) deposition because all forms can contribute to both acidification and nutrient enrichment of ecosystems. Moreover, the science indicates that ecological effects could be ameliorated by decreasing deposition of any of those forms. From a scientific standpoint, this intricate relationship indicates that the applicable NAAQS standard(s) should address total reactive nitrogen deposition. Thus, the Panel affirms that science-based secondary NAAQS standards for nitrogen and sulfur should consider the ecological consequences of total Nr deposition. However, it is noted that the present interpretation of the regulatory authority of EPA recognizes only oxidized forms of nitrogen and sulfur as Criteria Pollutants. The Panel recommends that the EPA move forward with establishing scientifically-based standards that effectively protect the environment, while recognizing the current regulatory constraints.

Scope of the Review

1. *Chapters 1 and 2 provide the background, history, and framework for this review, including a discussion of our focus on the four key ecological effect areas (aquatic acidification, terrestrial acidification, aquatic nutrient enrichment, terrestrial nutrient enrichment). Is this review appropriately focused in terms of characterizing the important atmospheric and ecologic variables that influence the deposition and, ultimately, the ecologic impacts of nitrogen and sulfur? Does the Panel have any further suggested refinements at this time?*

Chapters 1 and 2 of the REA begin to satisfy the goals of providing a clear and correct framing of the issues, providing a coherent method for reaching conclusions required in the NAAQS review process, summarizing the relevant policy questions and building on the ISA. The appropriate effects are considered in the REA and the potential relationships between atmospheric deposition and ecological effects are outlined.

The Panel commends EPA on its use of figures and diagrams throughout Chapters 1 and 2: such illustrations help orient the reader and provide a framework for discussion. It would be helpful if Figure 1.4-1 was accompanied by descriptions in the text that explain how well each of the steps can be executed, what the major uncertainties are, and where each component is addressed in the later parts of the REA. Such modifications would assist in guiding the reader through the material in the document. It would also be beneficial for the Agency to include a paragraph on the central concept of N and S “loading" because this concept is pivotal to the document. Similarly, the concept of “N saturation” needs to be introduced earlier in the document with a clear delineation of how this concept is, or should be, used in evaluating the effects of N loadings. There should be greater coherence between the text, figures and policy statements with respect to reduced and organic forms of N: both forms of N are mentioned here and in the ISA, and are important contributors to the effects being exhibited by ecosystems in response to N deposition. In the present version of the REA, the chemistry figures and the policy questions essentially address only the oxidized form. The material should be modified to address chemically-reduced and organic nitrogen.

Discussions of uncertainty in Section 2.4 are generic and qualitative: it is unclear when uncertainty is being discussed and when variability is the real issue. The document needs to be explicit that variability and uncertainty are two important but different concepts, with different impacts on establishing secondary NAAQS. The issue of uncertainty needs to be paramount in the discussion of monitoring networks. The sparse data from the existing network, the relationship between ambient concentrations and dry deposition and our understanding of cloud water deposition introduce significant uncertainty in estimates of total deposition. The Panel recommends that Chapters 1 or 2 address uncertainty with a discussion of the implications on EPA's ability to perform the assessments outlined in Chapters 7 and 8. Additionally, the uncertainty issue should extend to the discussion of climate change, for which there are both beneficial and adverse effects of atmospheric Nr in ambient air and deposition. The REA is unclear on how a change in the secondary NAAQS will affect climate change and in what direction. If the science is uncertain on the level and direction of effects on climate, it should be clearly stated in the REA.

Exclusion of non-ecological public welfare effects of NO_x and SO_x (e.g., effects of N- and S-containing particulate matter on visibility degradation and climate change, materials damage) continues to be an area of concern for the Panel. To ensure that the current REA meaningfully represents an assessment of environmental criteria, EPA should state clearly in which Agency documents the omitted NO_x/SO_x welfare effects will be treated and provide a short synopsis of the key non-deposition effects in appropriate locations in the REA.

The Panel recognizes that including a discussion of the potential primary NAAQS (i.e., based on public health) for NO_x and SO_x, and the current PM NAAQS, is premature at present. However, future drafts of the REA should include an analysis of how the estimates of ecological exposure and risk are impacted by the range of primary standards under consideration and if the respective primary NAAQS would (or would not) be adequately protective of the full range of environmental risks. Of particular concern is that in the past, the secondary NAAQS for a number of pollutants has been set identical to the primary NAAQS. As noted later in this letter, the Panel finds the proposed approach for a secondary NO_x-SO_x NAAQS to be innovative, but has concerns about its feasibility. Given the challenges the Agency may face with rulemaking and implementation, a policy decision could be to once again set the secondary NO_x and SO_x NAAQS to be identical to their respective primary NAAQS. We recommend a full assessment of the implications of making such a decision.

Air Quality Analyses

1. *To what extent are air quality characterizations and analyses presented in Chapter 3 technically sound, clearly communicated, appropriately characterized, and relevant to the review of the secondary NAAQS for NO_x and SO_x?*

2. *Section 3.2.1 describes an approach for evaluating the spatial and temporal patterns for nitrogen and sulfur deposition and associated ambient concentrations in the case study locations. This draft document includes the analysis for the Adirondacks case study. Does the Panel agree with this approach and should it be applied to the other Case Study Areas?*

3. *Section 3.2.2 describes the relative contributions of ambient emissions of nitrogen and ammonia to nitrogen deposition for the case study areas. To what extent is the approach taken technically sound, clearly communicated, and appropriately characterized?*

The general nature of Question 1 leads the Panel to both address the question and also to provide general comments on the current air quality analyses. Assuming the many and important missing "placeholder" sections are satisfactorily completed, the air quality characterizations and analyses presented in Chapter 3 should provide a reasonably sound and clearly communicated characterization of estimates of deposition of total reactive nitrogen and acidifying species to the specific case study areas. The Chapter, when completed, should also provide information that is relevant to potential revisions to the current secondary NAAQS for NO_x and SO_x.

Given that most of Chapter 3 is devoted to the presentation of model results from the Community Multiscale Air Quality (CMAQ) model and Response-Surface Model (RSM), a major limitation of the current chapter is the absence of any evaluation of model performance on simulated levels and atmospheric deposition rates of the various forms and phases of sulfur- and Nr-containing chemical species. It is critical that EPA complete the placeholders that relate to model/measurement comparisons and the characterizations of uncertainty in model results. For example, plots of spatial and temporal variability with measurements would convey how well the model captures the spatial and temporal patterns. Per the panel's previous recommendations on the ISA, much of the needed evaluation of the CMAQ model should be included in the ISA. There is also a need – either in the ISA or here in the REA – for a more detailed description of the RSM and an evaluation of its performance for the S and N species. With the relatively extensive sets of air quality, atmospheric deposition and environmental measurements available for the Adirondacks, this may be a good case study area in which to evaluate model performance prior to extending this approach to other case study areas.

The complex terrain in sections of the Adirondacks also raises concerns. It is not appropriate to compare the 12-km spatial resolution of the modeled deposition with the spatial patterns in actual deposition (including orographic precipitation increases and cloud water deposition) or the spatial patterns in sensitive species and ecosystems. A sensitivity analysis of variations within selected Adirondack model grid cells could help evaluate the importance of spatial variability, scale and resultant effects on deposition estimates.

The Panel was pleased to note the efforts of the Agency to link the atmospheric models (relating emissions to deposition) with watershed or landscape models (relating deposition to environmental effects). The disparity in the time scales between atmospheric (i.e., CMAQ) and watershed models is a major issue. Atmospheric models typically provide predictions at relatively fine time scales (e.g., hourly) over a single year, while the watershed models often provide results on scales ranging from decades to centuries. To the extent possible, it would be helpful if CMAQ could be run for a number of recent years (2002-2006), varying both meteorology and emissions, to provide a better understanding of inter-annual variability and longer-term spatial patterns. This would provide a more robust basis for model-to-measurement and model-to-model comparisons.

Assuming the model evaluations indicate reasonable performance for most of the chemical species of concern, the Panel recommends that the model results (for both sulfur and reactive nitrogen species) be used to generate a number of maps that illustrate the spatial distributions of:

- emissions (preferably interpolated to better illustrate high-emitting regions),
- atmospheric concentrations (gaseous and aerosol and combined species),
- deposition (wet, dry, total S, total N, oxidized N, chemically-reduced N, etc.), and
- ratios of deposition to atmospheric concentrations and deposition to emissions.

The Panel recommends that the maps have a consistent spatial resolution (e.g., 36 km) over the contiguous United States and a finer resolution over the case study areas. Such maps would provide the reader a more direct understanding of the spatial relationships that exist between emission location, ambient concentrations and the resulting deposition. The maps would also be useful in proposing a structure for a secondary NAAQS.

In addition to these maps, the Panel recommends the EPA use scatter plots of gridded model results to further illustrate the key relationships between the deposition metrics most relevant to environmental effects and alternative air quality metrics upon which the secondary NAAQS might be based. The scatter plots could also be used to provide information relevant to uncertainty analysis.

Case Study Analyses

Before dealing with the specific charge questions with regard to the Case Study Analyses, the Panel offers the following general comments. The proposed use of ecological indicators that can be linked to varying severity or magnitudes of effects (and related losses in ecosystem services) goes beyond the concept of estimating critical loads at which ecosystems experience no effects according to present knowledge. An important consideration as these indicators are selected and further developed is how to describe effect indicators of varying severity rather than just providing a "no-effects" threshold. A continuum of effect indicators will provide important information on the range of adverse responses, and quantifying the response indicators to various emissions and deposition levels will be important for the eventual assessment of what ecological effects are considered to be adverse. A separate section may be needed to discuss the ecological implications of various levels of acid neutralizing capacity (ANC) and associated effects, and its application to evaluating effects of acidifying deposition. This discussion should include a consideration of the implications for moving beyond single critical load “no-effects threshold” approaches to more comprehensive approaches that offer varying degrees of ecosystem protection within specified levels of statistical probability.

The modeling approaches used to develop critical loads for these case studies are very different. Evaluating these different approaches could be very instructive. In particular, a discussion of the relative merits of dynamic models vs. steady state models in these specific applications is needed. For example, MAGIC does not effectively simulate watershed nitrogen dynamics, so if watershed nitrogen dynamics is an important component of the critical load there will undoubtedly be some errors. Most forest ecosystems currently are losing exchangeable cations and accumulating S and N, so by definition they are not steady-state systems. Applying a steady-state model to such systems is problematic as a critical loads assessment tool.

In the case study Attachments, the discussions of varying amounts of ANC and their associated effects on ecosystem function differ from what is presented in the main chapters of the REA document. A more consistent approach is needed for setting the ANC limits of concern with respect to ecosystem sensitivity to acidification and recovery from acidification. The panel suggests that full protection of fish species (set at 100 μeq/L) should be considered in the case study scenarios.

It would be useful and instructive to include an N-limited site among the case studies. Possible systems would be the rapidly-growing and N-demanding Douglas-fir forest ecosystem in the Pacific Northwest or a loblolly pine dominated forest stands in the Southeastern US. For better understanding of the regional effects of N deposition in the southern Sierra Nevada, EPA should consider a comparison of responses in mixed conifer forests (e.g., the Kings River Project area) as compared to ANC changes in the nearby lakes of the sub-alpine and alpine zone (e.g., such as leaching of nitrate to streams, lichen species composition, and invasion of invasive grasses).

Additionally, it may be useful to include an up-front discussion of how the acidity of the solid phase of the soil affects soil solution acidity which can in turn affect surface water acidity. In an already acid soil (whether acidic due to natural or anthropogenic processes), soil solution acidity can increase or decrease instantaneously with the introduction or removal of mineral acid anions (such as sulfate, nitrate, or chloride) without any change in the soil solid phase. This is sometimes referred to as the "intensity effect" and is described in Reuss and Johnson (1986)[1]. In contrast, acidification of the solid phase of the soil can take a long time and is usually not reversible without liming.

1. *Attachment 2 presents a GIS analysis to define geographical areas that are sensitive to acidification and nutrient enrichment. Are the national geospatial datasets chosen adequate to identify sensitive areas? Are there other data sets that have not been identified by this analysis that we should consider? Does the panel agree with this approach or can they suggest alternatives?*

The selected datasets and the general GIS approach are appropriate for this analysis, and the Panel has no recommendation for additional datasets to consider. The period of coverage and spatial representativeness (e.g., the lichen database) are possibly important limitations that should be noted.

2. *Attachment 3 presents our current progress on evaluating the effect of aquatic acidification in the Adirondacks. It describes the use of the MAGIC model to evaluate ANC levels in selected streams and lakes in Adirondacks and Shenandoahs. To what extent is the approach taken technically sound, clearly communicated, and appropriately characterized?*

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1 Reuss J.O., and D.W. Johnson. 1986. Acid Deposition and the Acidification of Soil and Water. Chapter 7.5: Capacity versus Intensity. Ecological Studies No. 59. Springer-Verlag, New York, Pages 71-72.)

The selection of the Adirondacks and the Shenandoah Park as case study areas and the use of MAGIC as the modeling tool are appropriate; however, this section needs to be edited for clarity. At present, it is generally confusing, especially the modeling approach and descriptions of MAGIC and ASTRAP models. Numerous errors make it difficult to fully assess technical merits of the discussion in Attachment 3.

ANC has been selected as a metric to quantify the current acidic conditions and biological impacts because, in many studies, it was found to be the most appropriate single indicator of the biological response of aquatic communities in the acid-sensitive ecosystems and it is relatively easy to simulate using watershed biogeochemical models. Based on the ANC values and fish populations responses, five classes of biological responses (acute, severe, elevated, moderate and low concerns) have been developed and can by used for evaluation of risk assessment using the critical loads concept. As suggested above, for a higher level of protection of fish species EPA should consider using a more conservative value of 100 μeq/L instead of 50 μeq/L.

Some comparisons of these case study results with other efforts to evaluate acid deposition/biogeochemical responses in each of these sites (especially the Adirondacks) would be useful to indicate either differences from or support for these modeling efforts.

3. *Attachment 4 presents our current progress on evaluating the effect of terrestrial acidification. It outlines a plan to use the Simple Mass Balance Model to evaluate current deposition on forest soil ANC for sugar maple in Kane Experimental Forest and red spruce in Hubbard Brook Experimental Forest. To what extent is the approach taken technically sound, clearly communicated, and appropriately characterized?*

The selection of the study area is reasonable. Attachment 4 is highly uneven. Substantial detail has been provided in some cases, while broad generalizations are made in others. It is difficult for the reader to ascertain the salient points in this chapter.

4. *Attachment 5 presents our current progress on evaluating the effect of aquatic nutrient enrichment. It outlines a plan to evaluate how changes in nitrogen deposition affect the eutrophication index in two estuaries: Chesapeake Bay and Pamlico Sound. The analysis will model one steam reach (Potomac River and Neuse River) to determine the impact on the eutrophication index for the estuary. To what extent is the approach taken technically sound, clearly communicated, and appropriately characterized?*

The approach is appropriately characterized. The Chesapeake Bay has been the focus of considerable efforts relating to the effects of nitrogen loading on eutrophication. Extensive investigations have been made to identify Nr sources within both the very large airshed and the much smaller watershed areas of the Chesapeake region. However, a potential drawback of the choice of both the Chesapeake Bay and Pamlico Sound areas is that for both areas atmospheric nitrogen deposition is not likely to be the dominant component of the total nitrogen loading, and thus may not be as sensitive to atmospheric deposition changes as other ecosystems where deposition is the major source of nitrogen loading. Future assessments should consider including case studies of watersheds where atmospheric deposition provides a greater proportion of the total N loading.

5. *Attachment 6 presents our current progress on evaluating the effects of terrestrial nutrient enrichment. It describes an approach to evaluate the effects of N deposition on the Coastal Sage Scrub community in California and mixed conifer forests in San Bernardino and Sierra Nevada Mountains. To what extent is the approach taken technically sound, clearly communicated, and appropriately characterized?*

The selection of coastal sage scrub (CSS) and mixed conifer forest ecosystems is appropriate. Attachment 6 provides a comprehensive review of existing scientific knowledge for these two ecosystems and the findings have been clearly communicated. For development of critical loads for CSS, accumulation of biomass of invasive grasses (critical level for occurrence of catastrophic fires) could be considered. For the mixed conifer forests, change in lichen communities is a good indicator for ecosystem effects at the low end and nitrate leaching to surface water is a good indicator at the high end of total Nr deposition loads.

Additional Effects

1. *In this chapter, we have presented results from some initial qualitative analyses for additional effects including the impact of sulfur deposition on mercury methylation, the impact of nitrous oxide on climate change, and the impact of nitrogen deposition on carbon sequestration. Are these effects sufficiently addressed in light of the focus of this review on the other targeted effects in terms of available date to analyze them?*

These descriptions and level of detail in Chapter 6 seem adequate, but the relevant issues go beyond those associated with carbon sequestration, mercury methylation and nitrous oxide emission changes. The chapter would be much improved by providing a short summary that describes additional welfare effects of SO_x and NO_x for which deleterious impacts are expected (e.g., effects on visibility, climate and materials; as well as the deposition-related terrestrial and aquatic effects on which the REA currently focuses). Additional information is needed as to the effect of changes in pH on mercury concentrations in fish and other organisms. This summary should indicate both the importance of these ecosystems to individual regions and, in the context of the entire United States, the overall adverse ecological impacts. In considering the impacts of nitrogen deposition on carbon sequestration, the positive (e.g., increased forest growth and harvestable timber production) and the negative relationships need to be considered and presented in a balanced fashion.

Synthesis and Integration of the Case Study Results into the Standard Setting Process

1. *The purpose of Chapter 7 is to summarize the Case Study results and characterize the relationship between levels of an ecological indicator and the associated degree of ecologically adverse effects. To what extent is this approach characterized at this point of the review? Does the Panel have any further suggested refinements at this time?*

Chapter 7 remains in a very early stage of development; thus it is difficult to provide summary comments on the overall content and direction of this chapter. The Chapter ends abruptly and seems incomplete. Section 7.3 should be completed and a discussion of linkages between ecological indicators and adverse effects in terrestrial ecosystems is needed (similar to section 7.3.1 focusing on ANC in freshwater systems).

In general terms, the outline and material included in the Chapter are appropriate to the intended summary. Table 7.1 will be very important in its final form in future drafts of the REA and thus EPA should carefully consider its contents. The initial statement at the beginning of Section 7.2 strikes the right balance between important effects while recognizing their variable and localized nature. As appropriate throughout Chapter 7 (and the entire REA), the wording should be scrutinized so that the reader is clear when NO_x and SO_x versus total S or N deposition are the real drivers for the effects being considered. In the summarized cases, the key metric is total deposition irrespective of the original chemical forms of the airborne S and N inputs.

Considerations in the Structure of the NOx/SOx Secondary Standard

1. *Chapter 8 begins to explore how a secondary NAAQS might be structured to address the targeted ecological effects discussed in the risk assessment. The next draft of this document will include one or more examples of how this structure might be used to relate specific levels of air quality indicators with a corresponding ecological indicator for a given location and/or scenario. To what extent is the described approach technically sound, clearly communicated and appropriately characterized at this point of the review? Does the Panel have any further suggested refinements at this time?*

As noted previously, the Panel views that, scientifically, a NAAQS standard developed to protect ecological systems should focus on total acidifying deposition and excess nutrient enrichment, both of which include chemically-reduced, organic and oxidized forms of total reactive nitrogen. The current constraints have led EPA to focus on developing a combined ambient standard that is limited to sulfur and nitrogen oxides, but which may accommodate indirect consideration of contributions from other forms of reactive nitrogen as well. The proposed approach is scientifically well-founded: it is designed to lead to atmospheric deposition rates that will protect at least some (and perhaps many) of the target ecosystems as measured by specific ecosystem-based indicators of adverse ecological effects (e.g., ANC). While the approach does not directly include targeted decreases in atmospheric deposition of reduced or organic forms of nitrogen, the approach does include consideration of chemically reduced and organic nitrogen loadings as part of the already existing conditions that contribute to acidification and nutrient enrichment of sensitive ecosystems. As such, the approach could provide for a substantial decrease in some of the adverse ecological effects of acidification and nutrient enrichment in various part of the United States.

The Panel concludes that the approach presented is novel and environmentally relevant. The description should be clarified and more detail should be provided, specifically with regard to how the approach could be implemented in some of the proposed Case Study Areas. EPA should clarify how the approach would promote controlling chemically-reduced and oxidized nitrogen loadings as a means to decreasing ecological acidification and nutrient enrichment.

Currently, the discussion in Chapter 8 on establishing an appropriate linkage between ambient air concentrations and ecosystem effects, and the importance of spatial and temporal scales, is limited. The Chapter should show how the proposed approach directly addresses the Panel's concerns that, from a scientific perspective, the resulting environmentally-focused standard would include all reactive N deposition. Further, the Panel has concerns about the feasibility of implementing a standard based on this approach. There are a number of complications that need to be addressed so as to fully inform policy-makers, particularly in preparation for the development of an appropriate Advanced Notice of Proposed Rule Making (ANPR). Critical issues that need to be elaborated upon include:

1. whether (or how) a standard will integrate the multiple indicators identified in the case studies,
2. what are the appropriate spatial scales for each indicator and sensitive ecosystem,
3. how varying levels of protection required by different ecosystems can be accommodated, and
4. what level of protection is being provided to various ecosystems under alternative levels and forms of the standards.

The REA as a whole needs to show how the results of the case studies (specifically, the relationships between the observed chemical and biological changes and N and S atmospheric deposition) can be linked to the traditional NO_x, SO_x and PM standards that are based on ambient concentrations. It is imperative that this additional linkage information is incorporated in the next draft of Chapter 8.

In summary, the CASAC Panel was pleased to review this first draft of the *Risk and Exposure Assessment to Support the Review of the Secondary National Ambient Air Quality Standard for Oxides of Nitrogen and Sulfur*. The Agency's venture into new territory in the consideration of multi-pollutant standards is laudable. Shifting to standards that focus on ecological effects and employ metrics that are specifically relevant to ecosystems will have some inherent complexities and difficulties, and the Panel looks forward to following and contributing to the evolution of suggested approaches.

Sincerely,

/Signed/

Dr. Armistead (Ted) Russell, Chair
CASAC NO_x & SO_x Secondary
NAAQS Review Panel

/Signed/

Dr. Jonathan M. Samet, Chair
Clean Air Scientific Advisory Committee

Enclosures

Enclosure A

U.S. Environmental Protection Agency
Clean Air Scientific Advisory Committee
NO_x & SO_x Secondary NAAQS Review Panel

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Enclosure B

U.S. Environmental Protection Agency
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Enclosure C

<u>Comments received:</u>

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Dr. Praveen Amar

The charge is to respond to Questions #2 and 3 related to air quality analyses. Specifically, the two questions are reproduced here:

Question # 2 : Section 3.2.1 describes an approach for evaluating the spatial and temporal patterns for nitrogen and sulfur deposition and associated ambient concentrations in the case study locations. This draft document includes the analysis for the Adirondacks Case Study. *Does the Panel agree with this approach and should it be applied to the other Case- Study Areas?*

Question # 3 : Section 3.2.2 describes the relative contributions of ambient emissions of nitrogen and ammonia to nitrogen deposition for the case-study areas. *To what extent is the approach taken technically sound, clearly communicated, and appropriately characterized?*

Question # 2 Response:

The approach outlined in Section 3.2.1 should prove to be useful in that it does propose to do a complete analysis of spatial and temporal patterns of concentrations and deposition of sulfur and nitrogen compounds (dry oxidized nitrogen, dry reduced nitrogen, wet oxidized nitrogen, wet reduced nitrogen, dry sulfur deposition and wet sulfur deposition). However, the analyses at the present time are based only on CMAQ predictions (text says "CMAQ data"; "CMAQ predictions" is more appropriate) are only for one year (2002), and for just one case-study location (Adirondacks).

1. At a minimum, before one can answer the question "Does the Panel agree with this approach and should it be applied to other case-study areas?" with a reasonable level of confidence, the proposed approach needs to include an independent as well as corroborative (by comparing it to model-predicted results) analysis that is based on *measured* data for this case-study region (as well as the remaining case-study regions). It appears that modeled CMAQ results are reasonable, but it will increase the confidence in this approach if the measured data from NADP and CASTNet (and other networks in the Adirondacks region and other regions) corroborate the modeled predictions.

2. It is also important that before this approach is applied to other case-study areas, that the placeholders on Page 3-53 and 3-54 be completed. I would recommend that the analysis of inter-annual variation in N and S (for the years 2002-2006) deposition as well as uncertainty analysis (Section 3.2.1.5) be first completed for Adirondacks region before similar analyses are done for the other four case-study regions.

3. Once the measured deposition data analyses are completed, Section 3.2.1 should include a brief evaluation/comparison of CMAQ predictions for the four nitrogen and two sulfur components. As a part of this evaluation, the measured precipitation data and modeled (from MM5?) precipitation data (amounts and spatial patterns) should be compared. This

is important since the modeled results in Section 3.2.1 indicate strong correlations between amount of precipitation and wet deposition.

4. A general comment on presentation of results on dry deposition of N and S: This section needs to be more clear and explicit that we only ***estimate*** dry deposition (whereas we measure wet deposition) and therefore conclusions on total deposition (wet and dry) and on the relative contribution of each pathway have a level of uncertainty that is hard to determine, but needs to be acknowledged (for example, in Section 3.2.1.5 on Uncertainty).

Question # 3 Response:

This question involves the description of relative contributions of ambient emissions of NOx and ammonia to deposition of nitrogen (total nitrogen deposition (TND), oxidized nitrogen deposition (OND), and reduced nitrogen deposition (RND)) for the eight case-study regions. It asks if the approach used is technically sound, clearly communicated, and appropriately characterized. Here are some comments:

1. The "model of the model" or the RSM (Response-Surface Model) applied to CMAQ needs a more friendly description on how it works. On Page 3-55, the text makes an effort but does not succeed in explaining what (and how) exactly RSM does. It appears that RSM is like an "instrumented CMAQ" model in that it "represents the outputs of the CMAQ model using statistical predictions." It is not clear to me what exactly these statistical predictions are. It might be useful to compare the "workings" of RSM with, say, Direct Decoupled Method (DDM) or other "process" models (that evaluate the relative contribution of various processes embedded in the model on model predictions). Has the RSM approach been applied by the general scientific and policy/regulatory communities outside the US EPA?

2. To the extent RSM is essentially based on the "brute-force" approach of "zeroing out" NOx or ammonia emissions (recognizing there are some residual emissions for NOx that include international sources and lightning, and, for NH3, they include international, non-anthropogenic and point source emissions), I am not sure this is the right approach to accurately answer Question # 3. Are there more appropriate approaches that do not "unduly stress" the CMAQ model that can better address this question of relative contributions?

3. It is not clear to me how the twelve "emission control factors" on Page 3-56 were actually applied in the model. Were the emissions zeroed out only for the case-study region or for the whole modeling domain?

Dr. Andrzej Bytnerowicz

Question 2 – Current progress on evaluation the effects of aquatic acidification in the Adirondacks and Shenandoah. To what extent is the approach taken technically sound, clearly communicated, and appropriately characterized?

Selection of the Adirondack Mountains and Shenandoah National Park for estimation of ecological effects and risks caused by acidifying deposition of N and S is well justified. The two areas experience high levels of deposition, are characterized by a high density of anthropogenically acidified lakes and streams, and there is a well documented record of chemical and biological changes from many studies with their results published in peer-reviewed literature.

The proposed approach is logical and technically sound. The wet deposition data comes from the NADP/NTN networks operational in the Adirondacks since 1978 and in the Shenandoah since 1980. Current conditions were evaluated by a 3 step process that assessed trends in surface water SO_4^{2-}, NO_3^- and ANC concentrations; the percent of watershed bodies that have different degree of acidity; and the percent of water bodies receiving N & S deposition above the harmful levels (exceedance of critical loads). Biological effects of acidity caused by atmospheric N & S deposition are measured at the individual level as fitness and at the community level as species richness and community structure. ANC has been selected as a metric to quantify the current acidic conditions and biological impacts because in many studies it was found to be the best single indicator of the biological response of aquatic communities in the acid-sensitive ecosystems. Relationship between ANC and number of fish species showed that at the ANC values of 50-100 μeq/L, species richness begins to decline. Based on the ANC values and fish populations responses, five classes of biological responses (acute, severe, elevated, moderate and low concerns) have been developed and can by used for evaluation of risk assessment using the critical loads concept.

It will be interesting to see complete results of the planned evaluations.

Specific comments:

Page 32, Figure 5.1-2 - why there is such a high difference in the "severe" category between the observed and MAGIC modeled outputs?

Page 33, Figure 5.1-3 – change ANC units to μeq/L. In the same figure – why not to use more conservative value of 100 μeq/L instead of 50 μeq/L as the threshold of protection?

Question 5 – Current progress on evaluating the effects of terrestrial nutrient enrichment for coastal sage scrub and mixed conifer forests of California. To what extent is the approach taken technically sound, clearly communicated, and appropriately characterized?

Selection of coastal sage scrub (CSS) and mixed conifer forest ecosystems is appropriate because: (a) these ecosystems have high geographic coverage and are located in the important wildland–urban interface I highly populated areas, (b) they encompass a strong gradient of N deposition from the low, background levels up to the highest levels recorded in the US, (c) these ecosystems have been investigated for a long time from a perspective of interactive effects of atmospheric deposition, climate change fire and other stressors, (d) results of these investigations are well documented in the peer-reviewed literature.

If the goal of this chapter was to review the current state of science for these two case studies, than the approach taken was technically sound and findings have been clearly communicated. However, if results of these two case studies were supposed to show how the relationships between the observed chemical and biological changes and N atmospheric deposition (possibly expressed as critical loads) could be linked to the concentration-based NOx/SOx standards, then the used approach was not inadequate and should be revised.

The chapter provides a comprehensive review of the existing scientific knowledge for these two case studies. GIS maps show modeled N deposition in portion of California encompassing the two selected ecosystems, CSS threat from fire, and the presence of acidophyte lichens. Three-dimensional maps illustrating loss of CSS in relation to different levels of fire threats or N deposition could greatly help in developing a probabilistic approach to the evaluation of N deposition risks to important California ecosystems.

For development of CL for CSS, in addition to biodiversity changes or changes in lichen communities, accumulation of biomass of invasive grasses (critical level for occurrence of catastrophic fires) could be considered (Anderson, 1982; Scifres and Hamilton, 1993; Gimeno et al., 2009; Minnich and Franco-Vizcaino, 2009). For more information on this subject please contact Drs. Richard Minnich and Edith Allen, UC Riverside. For the mixed conifer forests, on the low end of N deposition changes in lichen communities, and on its high end, nitrate leaching to surface water, are good end points for CL estimates. Other, such as possible changes in understory biodiversity changes could also be considered and explored.

Data on N deposition levels was obtained from the NADP and CASTNET networks and modeled N deposition distribution from the CMAQ model runs for 2002, which were based on the 12 km grids. As the authors of this analysis suggest, results from the 4 km grid would greatly improve accuracy of predicted relationships between N deposition and the biological effects.

Some data for the main drivers of N dry deposition in the San Bernardino Mountains, gaseous HNO_3 and NH_3 from passive samplers have already been published (Bytnerowicz et al, 2007). More results on distribution of N nitrogenous gases monitored with passive samplers in southern California and southern Sierra Nevada are currently being prepared for publication (Andrzej Bytnerowicz, unpublished).

Suggestion: For better understanding of regional (southern Sierra Nevada) effects of N deposition, comparison of responses in mixed conifer forests (Kings River Project) such as leaching of nitrate to streams, lichen species composition, and invasion of invasive grasses, could be compared to ANC changes in the nearby lakes of the sub-alpine and alpine zone.

Specific comments:

Page 13, 1st paragraph – also high levels of NH_3 and NH_4^+ are deposited to CSS of southern California.

Page 29, Figure 3.1-1. Scale for N deposition is too coarse – a bracket 6.83-70.04 kg N/ha/yr is not acceptable.

Page 33, 1st paragraph – bark beetle should be added as important stressor in the mixed conifer forest ecosystem.

Page 44 and 45, section 5.2 – for the mixed conifer forest also changes of species composition of the under story vascular plants should be considered.

Page 45, list of questions – in regard to responses of lichens to N deposition, effects of oxidized vs. reduced N should be considered. This may be of interest because there is a potential shift towards less reduced N due to the movement of dairy farms from the Los Angeles Basin to California Central Valley.

References:

Anderson, H. E. 1982. Aids to determining fuel models for estimating fire behavior. USDA Forest Service Intermountain Forest and Range Experiment Station, General Technical Report INT-122, Ogden, Utah. http://www.fs.fed.us/rm/pubs_int/int_gtr122.pdf

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Gimeno, B. S., Yuan, F., Fenn, M. E., Meixner, T. 2009. Management options for mitigating nitrogen (N) losses from N-saturated mixed-conifer forests in California. In: A. Bytnerowicz, M. Arbaugh, A. Riebau and C. Andersen (eds). Wildland Fires and Air Pollution, Developments in Environmental Sciences, Vol 8, Elsevier, Amsterdam, pp. 425-455.

Minnich, R. A., Franco-Viscaino, E. 2009. A probabilistic view of chaparral and forest fire regimes in southern California and northern Baja California. In: A. Bytnerowicz, M. Arbaugh, A. Riebau and C. Andersen (eds). Wildland Fires and Air Pollution, Developments in Environmental Sciences, Vol 8, Elsevier, Amsterdam, pp. 339-364.

Scifres, C.F., Hamilton, WT. 1993. Prescribed Burning for Brushland Management: The South Texas Example. Texas A & M University Press, College Station.

Ms. Lauraine Chestnut

Charge question 1: Scope of the review
There seems to be some ambiguity regarding use of the term "sensitive." It sometimes seems to mean an ecosystem that is vulnerable but not necessarily harmed at current or historic deposition rates, and other times it seems to mean that harm is occurring at current exposures. To me, the word sensitive fits better for the former than for the latter. For there to be harmful effects it would seem to require both sensitivity and exposure. Thus, the selected case studies are appropriately selected as not just sensitive, but currently being affected by N and/or S deposition.

Pages 2-7 to 2.8

Descriptions of ecosystem services reduced or degraded as a result of harmful effects on ecosystem functions are not only important inputs into economic valuation and cost-benefit analysis, they are important in helping policy makers and the public understand the significance of the effects on the ecosystems.

It is important to recognize that economic valuation is best considered relative to an alternative. In figure 2.3-2 it is unclear what an economic value to "maintain" an ecosystem would be without specifying what would happen if some action were not taken. There may be a value to prevent a specified amount of degradation or a value to obtain a specified improvement, but the value to simply "maintain" is probably ambiguous.

Page 2-13

This discussion of uncertainty is pretty weak. One important note is that variability is not the same thing as uncertainty. There may be a lot of variability in how different ecosystems respond to the same amount of N/S deposition but it may be able to specify this variability with a great deal of certainty.

It is important to acknowledge uncertainty, but a critical thing here is how to determine when there is enough known to be able to set reasonable standards. The uncertainty issue will need to be taken up again when the analysis is further along. A key question is whether there is enough confidence in the results that they are useful to assist policy decision-making. This requires more than just listing sources of uncertainty, but necessitates an assessment of the significance of the uncertainty and how it affects the results.

Case studies

The proposed use of ecological indicators that can be linked to varying levels of effects (and related losses in ecosystem services) goes beyond the idea of estimating critical loads at which ecosystems experience no effects. This does not come through in the current draft until Chapter 7, and is a missing perspective in the various case study appendices. An important consideration that has to be made as these indicators are selected is their ability to be linked to effects of varying severity rather than defining simply a "no effects" threshold, because this will be important information for the eventual assessment what effects are adverse.

The REA ultimately needs to describe the significance of the effects on ecosystem function and services at current levels of exposure and at alternative levels of exposure that might be achieved with alternative standards in the case study areas. Selecting any secondary standard probably requires more than a determination of a “safe” level because the standard needs to protect against adverse effects, not just any effects. The case study analyses seem to be headed in this direction and I look forward to seeing this further fleshed out in the second draft.

Chapter 8

An important issue is that is not yet addressed is how the analysis will deal with spatial scale in defining a potential standard. This is complicated by expected variability in ecological response to deposition in different locations even within a case study ecosystem. It is unlikely to be reasonable or even feasible to set a standard to protect the most sensitive ecosystems (or components of an ecosystem). The scale decisions can be somewhat analysis driven in that the results may show reasonable categories or groupings, such as X% of lakes in the Adirondacks that would have an ANC of 50 or 100 at various ambient co

Dr. Ellis B. Cowling

My individual comments on the August 2008 First External Review Draft of the Risk and Exposure Assessment (REA) for the Secondary National Ambient Air Quality Standards for Oxides of Nitrogen and Oxides of Sulfur are organized below in response to each of the several Charge Questions posed the Panel in preparation for the October 1-2, 2008 CASAC meeting. As you will see, somewhat more detailed attention has been given to the five Charge Questions on Case Study Analyses and the recently received Chapters 7 and 8 than to other parts of this Risk and Exposure Assessment as requested by the Chair.

Scope of the Review

2. **Chapters 1 and 2 provide the background, history, and framework for this review, including a discussion of our focus on the four key ecological effect areas (aquatic acidification, terrestrial acidification, aquatic nutrient enrichment, terrestrial nutrient enrichment). Is this review appropriately focused in terms of characterizing the important atmospheric and ecologic variables that influence the deposition and, ultimately, the ecologic impacts of nitrogen and sulfur? Does the Panel have any further suggested refinements at this time?**

My most serious reservation about the analysis framework for Chapters 1 and 2 is that chemically reduced forms of nitrogen (NHx), organic forms of nitrogen (NCx), and total reactive nitrogen (Nr) all are not included in the specific wording of any of the 20 policy-relevant questions that are said to constitute the framework for this review on the effects of nitrogen and sulfur pollution on acidification and nutrient enrichment of aquatic and terrestrial ecosystems in the US.

This reservation is surprising in view of the very comprehensive analysis regarding the importance of chemically reduced forms of reactive nitrogen contained in the Integrated Science Assessment (ISA) document and also in view of the October 31, 2007 *Resolution* from the Science Advisory Board's Integrated Nitrogen Committee (INC) which makes the following strong assertion from the INCs Committee's examination of much of the same body of evidence reviewed in the ISA:

> "The current air pollution indicator for oxides of nitrogen, NOx, is an inadequate measure of reactive nitrogen in the atmospheric environment. The SAB's Integrated Nitrogen Committee recommends that inorganic reduced nitrogen (ammonia plus ammonium) and total oxidized nitrogen, NOy, be monitored as indicators of total chemically reactive nitrogen."

Please note the roster of current members of the SAB's Integrated Nitrogen Committee whose collective competence with regard the public welfare and public health effects of reactive nitrogen is very similar to the collective competence of the CASAC Panel selected to review Secondary NAAQSfor Oxides of Nitrogen and Oxides of Sulfur as can be seen in the Determination Memo at http://yosemite.epa.gov/sab/sabproduct.nsf/02ad90b136fc21ef85256eba00436459/c83c30afa4656bea85256ea10047e1e1!OpenDocument&TableRow=2.1#2.

The conclusionary statements written in **bold-type** in Chapter 4 of the ISA indicate that:

"The evidence is sufficient to infer a causal relationship between "acidifying deposition" (which includes NHx, and NCx, as well as NOx) and the following adverse acidification effects:

a) "changes in biogeochemistry related to terrestrial ecosystems,"
b) "changes in terrestrial biota,"
c) "changes in biogeochemistry related to aquatic ecosysytems,"
d) "changes in aquatic biota."

Also, "The evidence is sufficient to infer a causal relationship between Nr (reactive nitrogen) deposition (which also includes NHx, NCx, and NOx) and the following additional ecologically adverse nutrient- enrichment effects:

e) "alteration of biogeochemical cycling of N in terrestrial ecosystems,"
f) "alteration of biogeochemical cycling of C in terrestrial ecosystems,"
g) "alteration of biogeochemical flux of N_2O in terrestrial ecosystems,"
h) "alteration of biogeochemical flux of CH_4 in terrestrial ecosystems,"
i) "alteration of species richness, species composition and biodiversity in terrestrial ecosystems,"
j) "alteration of the biogeochemical cycling of N,"
k) "alteration of the biogeochemical cycling of C,"
l) "alteration of N_2O flux in wetland ecosystems,"
m) "alteration of CH_4 flux in wetland ecosystems,"
n) "alteration of species richness, species composition and biodiversity in wetland ecosystems,"
o) "alteration of biogeochemical cycling of C in freshwater aquatic ecosystems,"
p) "alteration of species richness, species composition and biodiversity in freshwater aquatic ecosystems,"
q) "alteration of the biogeochemical cycling of N in estuarine aquatic ecosystems,"
r) "alteration of the biogeochemical cycling of C in estuarine aquatic ecosystems,"
s) "alteration of species richness, species composition and biodiversity in estuarine aquatic ecosystems,"

"The evidence is sufficient to infer a causal relationship between:

t) "exposure to NO, NO_2, and PAN and injury to vegetation" and
s) "exposure to HNO_3 and changes to vegetation."

On the basis of this substantial body of accumulated evidence, I recommend that a schematic diagram similar to Figure 1.3-1 be included in Chapter 1 of the Second Draft REA document to illustrate the "cycle of reactive, chemically **reduced** nitrogen species." I also recommend that:

a) Chemically reduced (NHx) and also organic forms (NCx) of total reactive nitrogen also be included among the nitrogen pollutants of concern in many of the 20 policy-relevant questions listed in Section 1.4 on pages 1-17 through I-20 in Chapter 1 of this First Draft REA, and
b) Appropriate answers about both chemically reduced (NHx) forms of total reactive nitrogen, and, if possible also organic forms (NCx) of total reactive Nitrogen (Nr), be presented in Chapters 2, 3, and 4 of the Second Draft REA when it is completed.

<u>Air Quality Analyses</u>

4. **To what extent are air quality characterizations and analyses presented in Chapter 3 technically sound, clearly communicated, appropriately characterized, and relevant to the review of the secondary NAAQS for NOx and SOx?**

With the exception of the major reservations stated in answer to the Charge Questions about the Scope of the Review, I believe that the analyses presented in Chapter 3 are technically sound, clearly communicated, and appropriately characterized.

5. **Section 3.2.1 describes an approach for evaluating the spatial and temporal patterns for nitrogen and sulfur deposition and associated ambient concentrations in the case study locations. This draft document includes the analysis for the Adirondacks case study. Does the Panel agree with this approach and should it be applied to the other Case Study Areas?**

The approach used for evaluating the spatial and temporal patters for of N and S deposition and associated ambient concentration seems very reasonable to me. This approach proved to be useful in the Adirondacks Case study and I expect it to be reasonable for other case studies as well.

6. **Section 3.2.2 describes the relative contributions of ambient emissions of nitrogen and ammonia to nitrogen deposition for the case study areas. To what extent is the approach taken technically sound, clearly communicated, and appropriately characterized?**

I presume from examining the figures and associated text for the data presented on pages 3-63 through 3-112 that this question should have read:

" Section 3.2.2 described the relative contributions of chemically oxidized and chemically reduced forms to total reactive nitrogen for the Case Study areas. To what extent is the approach taken technically sound, …etc."

On the assumption that my presumption is correct, I consider this combination of modeling and measurement approaches to be reasonable. But I must confess that it took me a very long time to finally understand the rationale behind the statement on lines 10-12 on page 3-13 that "In order to calculate total nitrogen (by which I suppose the author meant deposition of total reactive nitrogen) the two chemical species from the National Atmospheric Deposition Program (NADP) (i.e., NO3- and NH4+) were added together and then added to the total dry deposition values estimated from the Community Multiscale Air Quality model (CMAQ).

It also took me a very long time to understand what was meant by the term "zero-out of NOx emissions" as used in most of the figure captions on pages 3-63 through 3-112) and periodically in the associated text.

With regard to the questions of "clearly communicated and appropriately characterized" I offer the following comments:

1) What a delight it was to find the following firm statement on lines 15-20 on page 3-70:

"Figures 3.2-42 examines the relative impact of emissions on NH_3 of the deposition of total reactive nitrogen. Figure 3.2-42 shows that NH_3 emissions represent a significant contribution to total reactive nitrogen in most case study areas, although the impact varies by season and by area. The smallest impact of NH_3, 10% occurs in the Potomac case study area in February. The largest impact of NH_3, 73% occurs in the Neuse Case study in July. The Neuse case study has the largest overall impact of from NH_3 of any of the case study areas, across all four seasons."

2) On lines 6 and 7 also on page 3-63 (and another case on lines 9 and 10 on page 3-71) we find three very confusing sentences that reveal clearly why EPA's constant use of the terms "reduce," "reducing" and "reduction" is so often confusing and ambiguous:

   "One possibility is that reducing NOx reduces HNO_3, which limits ammonium nitrate (NH_4NO_3) formation (and for existing aerosol, a reduction in HNO_3 shifts the equilibrium towards the gas phase), thereby increasing the lifetime of NH_3. A net increase in NH_3/NH_4 results. Because the deposit velocity of NH_3 is much higher than the deposition of NH_4^+ aerosol, dry deposition of NH_x increases.

   The terms "reduce," "reducing" and "reduction" have both chemical and numerical meanings. Fortunately we have the unambiguous terms "decrease" and "decreasing" which have only a single (always numerical) meaning. So why not use the unambiguous term "decrease" instead of the word "reduce" when our intended meaning is numerical and thus reserve the term "reduce" for its chemical meaning?

   Is this what was meant by the sentence quoted above?

   "One possibility is that **decreasing** emissions of NOx **decreases** air concentrations of HNO_3, which limits ammonium nitrate (NH_4NO_3) formation (and for existing aerosol, a **decrease** in HNO_3 emissions shifts the equilibrium towards the gas phase), thereby increasing the atmospheric lifetime of gaseous NH_3. A net increase in the ratio of gaseous NH_3 to NH_4^+ aerosol in the atmosphere results. Because the deposit velocity of gaseous NH_3 is much larger than the deposition velocity of NH_4^+ aerosol, dry deposition of NH_x increases.

<u>Case Study Analyses</u>

1. **Attachment 2 presents a GIS analysis to define geographical areas that are sensitive to acidification and nutrient enrichment. Are the national geospatial data sets chosen adequate to identify sensitive areas? Are there other data sets that have not identified by this analysis that we should consider? Does the Panel agree with approach or can they suggest alternatives?**

I have only limited experience with the several data bases that were used in the GIS analysis used in an attempt to define geographic areas that are sensitive to acidification. Thus I have only limited professional experience on which to base a detailed judgment in response to this question.

Nevertheless, my general impression derived from study of the summary map on page 18 of Attachment 2, and my general awareness of soil, vegetation, surface and ground waters, and the topographical, meteorological, and climatic factors that are relevant to acidification and nutrient enrichment, lead me to conclude that the GIS approach used is generally sound. I know of no additional data sets that should be included in this analysis.

2. **Attachment 3 presents our current progress on evaluating the effect of aquatic acidification in the Adirondacks. It describes the use of the MAGIC model to evaluate ANC levels in selected lakes and streams in the Adirondacks and Shenandoah. To what extent is the approach taken technically sound, clearly communicated, and appropriately characterized?**

I have no direct experience on which to base an informed judgment in response to this question. However, several of my colleagues tell me that the MAGIC model is very appropriate for these kinds of analyses.

3. **Attachment 4 presents our current progress on evaluating the effect of terrestrial acidification. It outlines a plan to use the Simple Mass Balance Model to evaluate current deposition levels on forest soil ANC for sugar maple in the Kane Experimental Forest and red spruce in the Hubbard Brook Experimental Forest. To what extent is the approach taken technically sound, clearly communicated, and appropriately characterized?**

I have studied Attachment 4 with considerable care and consider that the approach taken so far (since this is still a work in progress) is technically sound, clearly communicated, and appropriately characterized.

4. **Attachment 5 presents our current progress on evaluating the effect of aquatic nutrient enrichment. It outlines a plan to evaluate how changes in nitrogen deposition affect the eutrophication index in two estuaries: the Chesapeake Bay and Pamlico Sound. The analysis will model one stream reach (Potomac River and Neuse River) to determine the impact on the eutrophication index for the estuary. To what extent is the approach taken technically sound, clearly communicated, and appropriately characterized?**

I have studied attachment 5 with reasonable care and conclude that the approach taken in this case also is technically sound, clearly communicated, and appropriately characterized.

5. **Attachment 6 presents our current progress on evaluating the effects of terrestrial nutrient enrichment. It describes an approach to evaluate the effects of nitrogen deposition on the Coast Sage Scrub community in California and in mixed conifer forests in the San Bernardino and Sierra Nevada Mountains. To what extent is the approach taken technically sound, clearly communicated, and appropriately characterized?**

I have no experience with any of the western state ecosystems that are discussed in Attachment 6. Thus I have no professional experience on which to base an informed response to this question.

Additional Effects

1. **In this chapter, we have presented results from some initial qualitative analyses for additional effects including the impact of sulfur deposition on mercury methylation, the impact of nitrous oxide on climate change, and the impact of nitrogen deposition on carbon sequestration. Are these effects sufficiently addressed in light of the focus of this review on the other targeted effects in terms of available date to analyze them?**

Chapter 6 provides a useful overview of the impact of sulfur deposition on methylation of mercury which seems to me to be very worthy of consideration with regard to setting appropriate limits on the amount of air emission of sulfur oxides that should be permitted in various regions of the US. I see little reason for worry about nutrient enrichment or acidification effects of carbon sequestration. I do believe, however, that nitrous oxide emissions should be incorporated in EPA's review of the NAAQS for nitrogen pollution and its effects on terrestrial and aquatic ecosystems.

Synthesis and Integration of Case Study Results into the Standard Setting Process (Chapter 7)

1. **The purpose of Chapter 7 is to summarize the Case Study results and characterize the relationship between levels of an ecological indicator and the associated degree of**

ecologically adverse effects. To what extent is this approach technically sound, clearly communicated and appropriately characterized at this point of the review? Does the Panel have any further suggested refinements at this time?

I believe that the attempt being made in Chapter 7 to building a scientifically sound linkage between carefully selected ecological indicators and the extent and magnitude of ecologically adverse effects is a very complicated but very desirable goal.

During my initial readings of Chapter 7, and even more Chapter 8 during the few days between their availability to CASAC on September 23 and the CASAC meeting on October 1 and 2, it appeared to me that EPA was deliberately trying to build a case for making a modest decrease in emissions of nitrogen oxides (NOx -- which is already recognized as a Criteria Pollutant) –and of course would help decrease the adverse acidification and nutrient enrichment effects of total acidifying deposition and of total reactive nitrogen -- without also having to develop the very challenging regulatory case for "listing" ammonia and ammonium ions" as Criteria Pollutants.

At that time, my "seat-of-the-pants" inclination was to believe that the extent of decrease in reactive nitrogen pollution loads that are needed to protect sensitive terrestrial and aquatic ecosystems of this country probably cannot be achieved without taking steps to also include significant decreases in the amounts of chemically reduced and perhaps also organic forms of total reactive nitrogen.

During the CASAC meeting in the Research Triangle Park on October 1 and 2, 2000, however, and also during the EPA Science Advisory Board's Integrated Nitrogen Committee (INC) Workshop held in Washington DC on October 29 and 30, 2008, I began to see several things much more clearly:

1) EPA already has full regulatory authority to limit air concentrations of oxides of nitrogen in order to protect sensitive ecological systems from adverse effects,

2) The regulatory and political obstacles that would have to be overcome in order to "list" ammonia and ammonium ion as Criteria Pollutants are extraordinarily large,

3) It would not be possible to meet the court-ordered regulatory deadlines for completion of the current NOx and SOx Secondary NAAQS Standard reviews if an attempt were made to overcome these very imposing regulatory and political obstacles,

4) It would be possible to consider the amounts of NHx and NCx as part of the "already existing conditions that lead to acidification and nutrient enrichment of sensitive aquatic and terrestrial ecosystems,"

5) It should be possible to determine what technically achievable extent of decrease in emissions of NOx might be sufficient to protect some if not all sensitive ecosystems in various parts of the US from the adverse ecological effects caused by reactive-nitrogen-induced acidification and nutrient enrichment of aquatic and terrestrial ecosystems, and

The present drafts of Chapters 7 and 8 indicate that OAQPS is seeking first to determine, within EPA's **present** regulatory structure, if decreases in emissions of nitrogen oxides (NOx) -- which are already recognized as a Criteria Pollutant -- could achieve an adequate decrease in the adverse acidification and nutrient enrichment effects of N pollution that are now occurring in various parts of the United States.

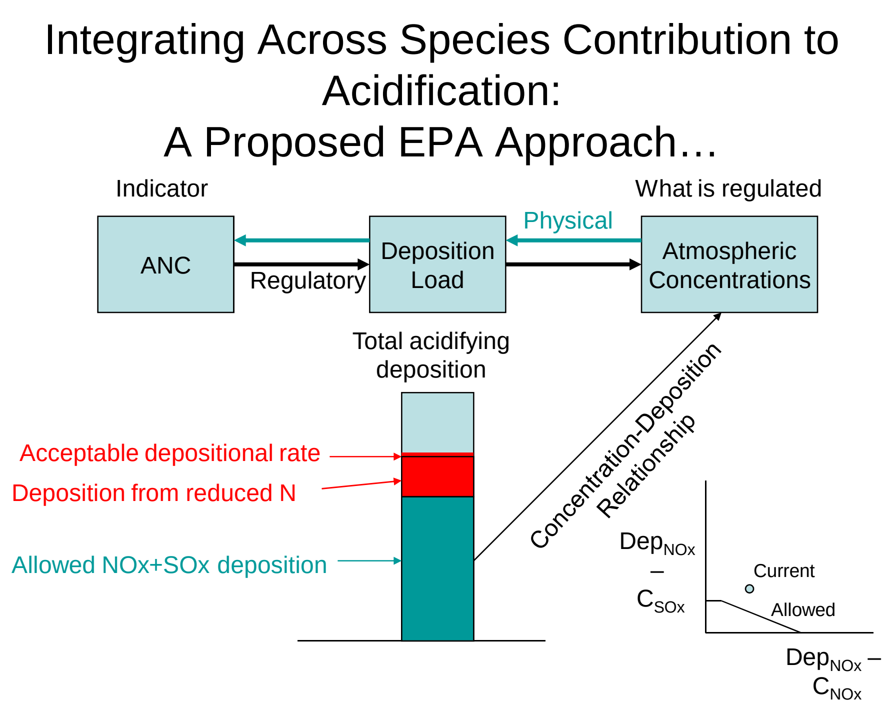
During our CASAC discussions with OAQPS on October 2, 2008, we were assured that Chapters 7 and 8 of the Second Draft REA will deal much more completely with the combined causal effects of both chemically reduced (NHx) and chemically oxidized forms of reactive nitrogen (NOx).

Considerations in the Structure of the NOx/SOx Secondary Standard (Chapter 8)

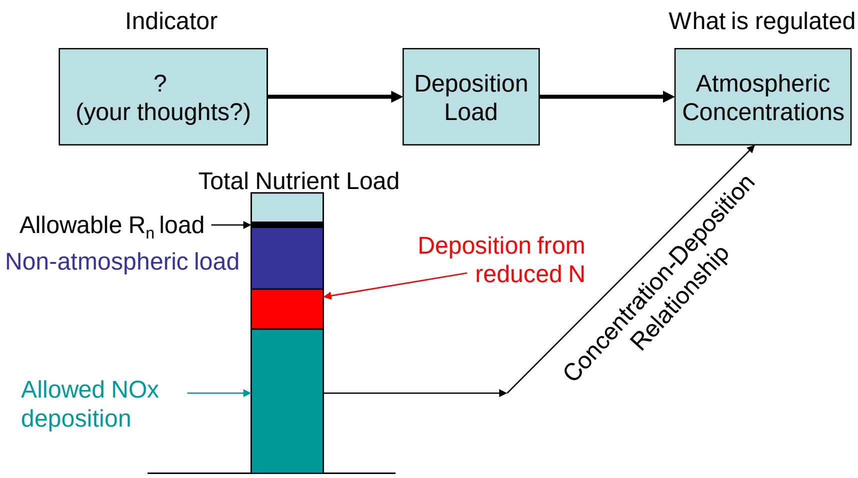
1. **Chapter 8 begins to explore how a secondary NAAQS might be structured to address the targeted ecological effects discussed in the assessment. The next draft of this document will include one or more examples of how this structure might be used to relate specific levels of air quality indicators with a corresponding ecological indicator for a given location and/or scenario. To What extent is the described approach technically sound, clearly communicated and appropriately characterized at this point of the review? Does the Panel have any further suggested refinements at this time?**

In Section 8.1 – *Possible Structure of a Secondary Standard* – the diagram in Figure 8.1-1 was especially informative of the approach that is currently being considered by OAQPS. The example given was based on the ecological indicator ANC (acid neutralizing capacity) and assumed that the pollutants of concern were not only the chemically oxides form of reactive nitrogen (NOx) and sulfur (SOx) , but also the chemically reduced forms of reactive nitrogen (NHx).

The attached diagrams were developed by Ted Russell in his role as Chair of the CASAC NOx/SOx Secondary Standards Review Panel for presentation to the SAB's INC Committee during the INC Workshop in Washington DC on October 30, 2008. These two diagrams were designed to show how OAQPS might include an estimate of chemically reduced nitrogen as one of the "already existing conditions" that lead to acidification and/or nutrient enrichment of aquatic and terrestrial ecosystems, and thus to provide a basis for calculating the amount by which nitrogen oxide deposition would need to be decreased in order to achieve an appropriate critical load for acidification effects and a similarly appropriate critical load for nutrient enrichment.



Applied to Nutrient Enrichment



Dr. Douglas Crawford-Brown

I am charged primarily with Scope of the Review, and so my comments are primarily on that issue. The specific Charge Question addressed is:

"Scope of the Review: Chapters 1 and 2 provide the background, history and framework for this review, including a discussion of our focus on the four ecological effect areas (aquatic acidification, terrestrial acidification, aquatic nutrient enrichment, terrestrial nutrient enrichment). Is this review appropriately focused in terms of characterizing the important atmospheric and ecologic variables that influence the deposition and ultimately the ecologic impacts of nitrogen and sulphur? Does the panel have any further suggested refinements at this time?"

This review also considers Chapters 7 and 8, as these are the core of the document in regards to an eventual regulatory decision.

As with my review of the ISA, my conclusion here is that the REA does in fact satisfy the goal in the Charge Question, subject to the comments below. The correct effects are considered (there may be more effects one could note, but the ones considered here are the most significant and are likely to bound the areas of concern adequately), and the correct relationships to atmospheric and ecologic variables are considered (again to the extent these are needed to draw the primary conclusions). The document is well written, being easy to follow and nicely organized, although the wheels fall off a bit – or are not even present - in Chapters 7 and 8). The authors have culled the most important conclusions from an immense literature, focusing the reader properly onto the key findings. A theme that will emerge below, however, is my feeling that the available data and analyses may support the need for considering a reduced NAAQS for NOx and SOx, but is insufficient to suggest the actual ambient levels needed to avoid demonstrably adverse effects (which I contrast with effects alone, which may or may not be sufficient to deem adverse).

This document would benefit greatly from an Executive Summary similar to the one in the ISA. There is a large amount of information here, but it can be boiled down to a few key conclusions. My fear is that failing to do that, the authors may find specific parts of the document picked in the policy process because they support a desired conclusion and policy solution. There needs to be a concise and unambiguous statement of the key scientific conclusions, and an Executive Summary is exactly the place to put these.

I found Chapter 1 very well written. The document lays out the relevant policy questions and even relates these (in contrast to past documents) clearly to the task of deciding whether the NAAQS needs to be revised and, if so, how the information would be used to do that.

Figure 1.4-1 is quite interesting, but it also lays clear the one glaring problem with a secondary NAAQS, since the key element is the Ecological Effect Function. I don't see where such a function is sufficiently well established to allow use in setting a secondary standard, other than perhaps as an analogue to an effects threshold in non-cancer human health risk assessment, probably with some margin of safety inherent in it due to the inability to draw a proper line between effect and adverse effect.

Chapter 2 begins by listing the appropriate effects, and I agree with the selection of these based on the information in the ISA. Table 2.1-1 is particularly useful in providing a road map to the material in the entire document. In the previous draft, I was unclear as to the purpose of the case studies. In the present draft, this point is clearer, and I agree with the idea that given the very large inhomogeneity in both exposure conditions and species across different geographic areas, the best that can be done is to select a few representative but sensitive regions and determine where the ambient levels would need to be to protect these. The one thing missing is a clear statement as to how unique these case study areas are. One can't set national standards based on a few outliers in the national distribution, and I believe a better case can be made in the document as to why these particular areas studied are not in the extremes of the tail of inter-site distributions of sensitivity.

I particularly like the structure of the assessment outlined in the seven steps. While the committee may have disagreements over specific methodological issues, these seem to me the appropriate steps and an innovative way to get at the issue of a secondary NAAQS that relies maximally on available data. I fully support the EPA staff in this choice of framework, even if in the end they must execute it somewhat more qualitatively than might be desired. The point is that it is the right way to be thinking about the NAAQS.

The ecosystem services discussion in Chapter 2 was interesting to read. It presented the subject well, and it is evident to me that ecosystem services is one lens through which to view a secondary NAAQS (although it doesn't capture issues such as inherent rights of other species). My problem lies in a disconnect between the detailed discussion of ecosystem services and the specific Ecological Effect Functions in Figure 1.4-1. I don't believe the document, or even the current state of the science, allows for development of such a Function needed to determine how much a specific ecosystem service is impacted by a given N or S loading, or how adverse is a given decline in ecosystem service. I wouldn't be inclined to support a position that says any decline is automatically adverse; the same applies to my position on human health imapcts. Due to this methodological and computational gap, the Ecosystem Services discussion in Chapter 2 comes off as more interesting than truly informative – a good idea that can't quite be pulled off when the data are analyzed.

The uncertainty discussion, as in almost all of the REAs we have reviewed, is quite generic and qualitative. But given the nature of this exercise, I am not sure a more quantitative approach to uncertainty would inform the final decision. This is because, while there are quantitative uncertainties having to do with the data and modelling, an equally important uncertainty is the conceptual relationship between the case studies and any sort of statement about the impact of a national standard.

As Chapters 3, 4 and 5 are outside my area of expertise, especially with respect to specifying where the staff should look for representative but sensitive case study areas, I don't provide comments here, other than to note that Chapters 4 and 5 are of little use given their sketchy nature.

The heart of the REA is found in Chapters 7 and 8. Table 7.1-1 agrees with the information provided in Chapter 1, so at least the methodology is consistent on this point. The framework of thinking laid out in this Chapter is appropriate, although provided here in much too sketchy a form for me to agree or disagree with how it is being executed. There remain two areas in which substantial disagreement can arise between individuals reviewing the document: (1) the methodological steps in calculating impacts on a given case study site and (2) drawing summary conclusions across sites. At the moment, the document does not fully clarify the first, and the second issue is dealt with more through aspirations than any clear approach. But I must withhold judgment until the final report is prepared. The staff is at least headed in the right direction, have a proper roadmap in front of them and have the expertise on hand to carry out these tasks.

In Chapter 8, the phrase "uniform level of ecosystem protection" occurs, and seems to become a key idea in how a NAAQS might be considered. This idea really needs more of an explanation. Given the high levels of inhomogeneity, and the fact that the conclusions rest ultimately on case studies of sensitive areas, and the quite diverse kinds of effects being considered, I don't understand what is meant by a "uniform level of ecosystem protection". It surely doesn't mean that the level of effect will be the same across all ecosystems in the country, or even that the same ambient level will produce the same level of effect everywhere, or that the effects will be equally adverse in some deeper sense. And there is no common metric to which all these diverse effects can be reduced. So, just what does it mean?

Again, Figure 8.1-1 is the right kind of structure, but I don't see how the Ecological Effect Function will be developed as anything other than a threshold model. And I don't see where a margin of safety is recognized or introduced. But it is still the right conceptual approach if it can be pulled off methodologically.

Much of the discussion in Section 8.2 seems to me of a policy nature, belonging in a much earlier chapter. It almost comes across as being filler here while the staff tries to figure out exactly how they will execute the ambitious steps in Figure 8.1-1. I recommend moving it to the front of the REA in either Chapter 1 or 2.

The rest of the Chapter 8 strikes me as a lot of scientific detail with little to connect it all to the final calculations. I can't comment on many of the equations proposed, because they relate more to environmental transport and fate than to effects. But it is evident to me that there is still a large gap between methodologies to estimate deposition and methodologies to relate these loadings to any specific effect that will drive a NAAQS.

Dr. Charles T. Driscoll

The document “Risk and Exposure Assessment for Review of Secondary National Ambient Air Quality Standards for Oxides of Nitrogen and Oxides of Sulfur” is an effort by U.S. EPA staff to provide and discuss a framework for establishing secondary standards of nitrogen oxides (NO_x) and sulfur dioxide (SO_2). Overall, I found the framework thought provoking and an interesting path forward in the establishment of secondary standards. While I enjoyed reviewing the document, there were several general technical issues that I am concerned about. There are several wording and grammatical problems in the text that should be addressed before the document is more widely circulated. Finally, there are numerous small technical and wording problems in the document. I have organized my comments around these three issues.

Unfortunately, the document is not complete. Major sections are missing or partially complete. It is really a waste of time to conduct a review of such a large and complex document when the document is incomplete.

Technical Issues:

1. *Case studies and modeling approach.* I think the approach of using case studies to address the framework for secondary standards is a good and appropriate one. I also generally think the specific case studies that are advanced in the REA are appropriate and helpful. I generally endorse the approach used. There are a few general comments/issues I would like to address.

   The case studies for aquatic effects in the Adirondacks and the Shenandoah Park regions, terrestrial effects on red spruce and sugar maple in the East and terrestrial effects on coastal sage scrub and conifer forests in California seem appropriate. I also like the two estuarine sites to evaluate coastal effects. I do have some concerns with the estuarine studies. First, it appears that the entire estuary watershed will not be evaluated (i.e., Chesapeake-Potomac; Pamlico; Neuse). Will this be a problem or is the scope of doing the entire watershed just too great for this assessment? More problematic is conducting analysis for two watersheds that are in fairly close proximity? Although there is considerable information for Chesapeake Bay and Pamlico Sound, wouldn’t it make more sense to conduct one of these case studies at a site with more contrasting features, with different land cover, climatic or N sources? I would think interest’s would be best served by either selecting a northern estuary (e.g., Gulf of Maine, Long Island Sound) or a Gulf estuary as a second site.

   The other general technical comment is that the approaches used to develop critical loads for these case studies are very different. Evaluating these different approaches could be very instructive but also problematic. I like the approach proposed of using a dynamic acidification model for the Adirondack and Shenandoah case studies. Note, however, that MAGIC does not effectively simulate watershed nitrogen dynamics. So if this is an important component of the critical load, there will undoubtedly be some errors. I find it disconcerting that for the terrestrial acidification assessment a steady-state model is being used. First, is it a good idea to use a dynamic model for the aquatic assessment and a steady-state model for the terrestrial assessment? While I agree with the authors that critical loads are a steady-state phenomenon, ecosystems are not. Forest ecosystems are losing exchangeable cations and presumably accumulating sulfur and nitrogen. This makes these systems by definition not a steady-state and increasingly sensitive to inputs of acidic deposition. While the application of a steady-state model is easy, it would seem to be problematic as an assessment tool. There are clear limitations in using a steady-state model for critical loads assessments. Finally, I don't understand how the critical loads will be determined for the coastal and terrestrial nutrient case studies. SPARROW is a statistical model and it is not clear how this can be used to evaluate greater and lower N loads.

2. *Nitrogen saturation.* Throughout the text, N saturation is referred to. However, I could not find any discussion of this phenomenon in the introductory sections. It is discussed in the ISA but not (that I could find) in the REA. A brief summary of N saturation might be helpful.

3. *Time scale disconnect.* There appears to be a disconnect between the time scales used for the atmospheric modeling and the effects assessments. Ecosystem effects of air pollutants are largely manifested over decades to multiple decades. Certainly the simulations conducted by MAGIC are conducted with what I believe to be the appropriate temporal perspective. I believe the time-scale for nutrient effects on ecosystems similarly have a long-term perspective. In contrast, the deposition/CMAQ analysis seems to be largely focused on a short-term or seasonal perspective. Why? There seems to be a complete disconnect in the atmospheric and effects modeling concerning time-scale of analysis. Isn't the primary concern here ecological effects? Do seasonal or monthly patterns in air concentration or deposition have any relevance for this long-term analysis of ecosystem effects?

4. *Climate.* I am a bit surprised that no discussion is given to changing climate. The framework to be developed is examining effects that will play out over the next decades. It is projected that climate will also change substantially over the same period. Climate change will affect hydrology and ecosystem response to air pollution. Climate change should be mentioned and needs to be addressed in future assessments.

5. *Establishing standards around ambient air concentrations.* In Chapter 8, limited discussion was advanced in establishing ecosystem effects around ambient air concentrations. While I can see that this might be a desirable objective, as we currently have primary standards and some quasi secondary standards based on ambient air concentrations. However, for ecosystem effects, I do not see this approach as workable. I think the standard needs to be based ultimately on total sulfur and total nitrogen deposition. There are many species of sulfur and nitrogen all which contribute to ecological effects but having different residence times in the atmosphere. These residence times vary in time and space. The key driver of ecological effects is long-term

total deposition. Establishing standards around ambient air concentrations would seem to be intractable.

6. *Spatial variability in sensitivity.* I'm not sure if this consideration is relevant for the nutrient case studies, however, the acidification case studies will exhibit considerable spatial variability in sensitivity to acidic deposition. There is a range of ecosystem sensitivity to acidification from highly sensitive to highly insensitive. How will this range of ecosystem response to acidic deposition be addressed when establishing the critical load? Will all ecosystems be protected? 90%, 50%. Some discussion of this consideration would be helpful.

Written Document Considerations:

1. *Written perspective.* The document is written from the "we" perspective (i.e., we did this…, we analyzed that…). I find this approach somewhat disconcerting. The reason being it is not clear who owns the document. Is this EPA's document or the contractor's document? Who are we? I would like to see the document altered.

2. *Typos, errors, writing mistakes.* As with the last draft of the ISA, the REA (and the 2nd draft of the ISA) is filled with mistakes and typos. I point out many of these in my specific comments (see below). However, these are by no means all the mistakes. This document needs to be carefully read, proofread and edited for consistency and to eliminate the mistakes.

3. *Redundancy.* There are many redundant sections in the document. This makes a very long document, longer than it needs to be. The document should be edited to eliminate the redundant text.

4. *Tense.* The REA switches back and forth from the past to present tenses. I can see writing in either tense. However, the document should be edited so it is written in a consistent tense.

Specific comments:

Page 1-2, line 16	Units of ANC μeq/L?
Page 1-11, line 24	I don't agree with the statement. Most published studies document inputs of oxidized and reduced N. A few don't, but most do.
Page 1-12, line 10	Space missing.
Page 1-12, line 13	I would change the wording. Acidification is an environmental effect due primarily to sulfur and secondarily nitrogen in most environments.

Page 1-13, line 12	As above, need to define N saturation.
Page 1-13, line 19	Change air to atmosphere.
Page 1-15, line 22	alpine
Page 1-16, line 12	It is incorrect to state that watersheds conducive to methylation are found in the northeastern U.S. and southeastern Canada. They are found all over. See Figure 6.1-3.
Page 2-2, table 2.1-2	Under aquatic acidification also include hydrologic flow paths under sensitivity variable.
Page 2-4, line 15	Eliminate comma
Page 3-7	I would like to see this section expanded to include a section of background (pre Industrial Revolution) deposition, including sulfate, nitrate, ammonium and basic cation deposition.
Page 3-8	I would like to see a brief description of organic N deposition, including sources.
Page 3-9, thru 12	There is a summary of spatial deposition patterns. The discussion of how these maps are produced is in the section that follows (3-12). The methods section should be moved in front of the maps. Also, the maps are generated for 2002. Some discussion should be given as how representative this year is, given year-to-year variability in deposition. Moreover, it is critical to clarify on the deposition maps the units of mass (e.g., Kg N/ha-yr or kg NO_3/ha-yr).
Page 3-12, line 10	data are …
Page 3-13, line 1	data were…
Page 3-13, line 6	Change to… kg/ha-yr.
Page 3-13, line 7	data were…
Page 3-16, line 5	Need to clarify the time interval kg/ha-yr?
Page 3-16, table 3.2-2	It is not clear what this table is. Some additional text is necessary.
Page 3-19	It would be helpful to put the Adirondack Park and the Shenandoah National Park boundaries and the Chesapeake and

Pamlico watershed areas on the map so the reader can understand the scope of the analysis relative to the total resource.

Page 3-25, line 7,8 — Change to… kg N-ha/yr.

Page 3-35, line 12 — Change to… fairly uniform.

Page 3-36, figure 3.2-14 — Why show figures of both monthly and seasonal deposition? Isn't this redundant?

Page 3-38, figure 3.2-16, 17, 18, 19
These figures are difficult to read and are they really helpful?

Page 3-40, line 4 — Change to… generally uniform…

Page 3-50, lines 3, 17 and Page 3-51, line 6
Change…drop to decrease

Page 3-50, figure 3.2-28 and elsewhere
Aren't the monthly patterns in wet deposition strongly driven by the quantity of precipitation? Wouldn't patterns for a different year with different precipitation patterns be different? As a result, this temporal section is misleading because it is strongly affected by the meteorology for that year. If true, why include all this analysis? As a minimum, this fact should be clarified and some data provided on 2002 as a reference year.

Page 6-1, line 3-4 — Rephrase. Every wetland has sulfate. The production of methyl mercury is largely mediated by sulfate reducing bacteria.

Page 6-1, line 22 — Also phosphorus (or N) can be important as it regulates aquatic productivity and therefore mercury concentrations in aquatic organisms (Driscoll et al. 2007).

Page 6-2, line 4 — Change to…Industrial Revolution.

Page 6-2, line 28 — This sentence needs to be clarified. Ionic mercury can be reduced and evaded and separately methylated. Methyl mercury is not reduced. Where does this 1-2% come from? The extent of methylation is highly variable from ecosystem to ecosystem.

Page 6-4, line 14-16 — Why is methane needed? Why do you need HgS in the equation? Define $MeHg^{+}$.

Page 6-4, line 20 — Also anoxic conditions.

Page 6-4, last paragraph	There are other studies that probably should be cited (Branfireun et al. 1999), (Jeremiason et al. 2006).
Page 6-5, line 12	Change to…methylation can occur within.
Page 6-5, figure 6.1-2	Hg can also be supplied from sediments.
Page 6-6, line 3	Change to… anoxia, sulfate).
Page 6-6, line 5	Change to… response, hydrology, nutrient loading, limnology).
Page 6-10, line 4	Is this really true? I do not believe it. % methyl mercury is highly variable. Need to correct.
Page 6-12, line 5	Change to… oxidizing NH_4^+.
Page A3-1, line 15	Subscript 2.
Page A3-1, line 20	hydrogen ion, and Al^{n+}.

Page A3-1, line 20 and throughout the document

I don't think the writers of this document understand the concept of buffering capacity. Buffering capacity is the resistance of a system to changes in pH. I would recommend eliminate using the term here and elsewhere or change the phrasing to use it correctly. An alternative could be acid neutralizing capacity or acid-base status.

Page A3-2, 1st paragraph	I suggest adding a sentence or two about immobilization/mobilization of SO_4^{2-} and NO_3^- by plants/soil organic matter.
Page A3-2, line 10	There are numerous chemical indicators.
Page A3-2, line 12	NO_3^-, Al^{n+}
Page A3-2, line12	Change to… of base cations; and ANC.
Page A3-2, line 27	Change to… precipitation enters the soil and soil water to emerge…
Page A3-2, line30	$K^+ + Na^+ + NH_4^+) – (SO_4^{2-} + NO_3^- + …)$ in (μeq/L)
Page A3-3, line 5	Change to… low pH.

Page A3-3, line 6	Change to… This is the acid neutralizing capacity (ANC), or the …
Page A3-5, line 4	acidic surface waters (14%; ANC<0 μeq/L).
Page A3-5, line 26	United States
Page A3-6, line 24	Need to define the time and mass basis of deposition (e.g., kg SO_4/ha-yr or kg S/ha-yr).
Page A3-7, line 12	Change to… weathering rates and limited neutralizing of acid inputs.
Page A3-8, line 5	Again need to define the time and mass basis of deposition.
Page A3-9, line 19	Al^{n+}
Page A3-9, line 21	with limited leaching
Page A3-9, line 23	EPA-administered Long-Term Monitoring (LTM) program.
Page A3-15, line 12	SO_4^{2-}
Page A3-16, line 9	comma
Page A3-16, line 19	it's the acid neutralizing capacity of a …
Page A3-16, line 21	The acid neutralizing capacity of a …
Page A3-17, line 3	20 μeq/L (limited protection)
Page A3-16, line 12	This sentence makes no sense. At ANC = 0 μeq/L a water is chronically acidic.
Page A3-17, line 8	Sub and super script
Page A3-20, line 5	Units should be eq/ha-yr.
Page A3-20, line 7,8	I would eliminate the term occult deposition simply call it cloud and fog deposition.
Page A3-20, line 10	Units meq/m^2-yr
Page A3-22, line 24	This description needs to be expanded or clarified. There are more than 200 NADP sites.

Page A3-28. line 25	in the catchment
Page A3-29, line 19	400 meq/m^3
Page A3-29, line 24	The titles used for these classes should be consistent with the titles established in table 4.1-1 on A-14.
Page A3-33	There needs to be some discussion on the time and nitrogen retention assumptions used to obtain critical loads.
Page A4-1	I would change the title to Forest Acidification Case Study.
Page A4-1, line 17	hydrogen ions
Page A4-1, line 19	where strong acids
Page A4-4	Should also consider citing the recent paper by (Warby et al. (in press)) attached), which shows widespread soil acidification in the Northeast.
Page A4-5, line 14	This statement needs to be reworded. Al mobilization occurs under low % base saturation and high concentrations of acid anions. The statement as it stands is incorrect.
Page A4-9, line 18	Should be (Driscoll et al. 2001).
Page A4-10	It would be helpful to cite the study by (St. Clair et al. 2005) which shows decreases in foliar antioxidant enzymes in sugar maple in response to lower foliar and soil Ca^{2+} in Pennsylvania.
Page A4-19, line 1	Again need to specify the mass and time basis of deposition.
Page A4-28, line 10	acidity input neutralized by
Page A4-28, line 18	parties
Page A4-49	Need to indicate the units of the figure.
Page A4 References	Should be BioScience.
Page A5-18, 1st paragraph	Need to use metric units.
Page A5-26, line 6	Need to define Nr.
Page A5-30	SPARROW is a steady-state model. Will need to demonstrate how you can use it.

Page A5-55, line 7	were from
Page A5-58, line 6, 8	Does the SAV coverage really have this level of significant figures?
Page A6-10, line 1	Change to… Mediterranean climate. This climate is…\
Page A6-25, line 16	result of long-term elevated N deposition rather than pulses
Page A6-42, line 7	data are
Page 7-3, line 12	Change to… where strong acids are
Page 7-11, line 2	catchment to neutralized acid anion deposition is known as acid neutralizing capacity.
Page 8-1, line 14	There cannot possibly be a uniform level of ecosystem protection due to the inherent variability in ecosystem sensitivity.
Page 8-4, figure 8.1-1	Add climate as a variable/fixed factor.
Page 8-5, line 2	What is meant by the point of deposition?
Page 8-8, line 14	What about forest acidification?
Page 8-8, line 14	How can you say whether precipitation occurs or not. Is there a location where precipitation does not occur? Rewrite sentence.
Page 8-10, line 24.	Do you mean deposition is expressed or an equivalence basis? Please clarify.
Page 8-11, line 4	This sentence makes no sense and should be rewritten.
Page 8-14, line 1	data are
Page 8-14, line 20	Do you mean equivalence ratio?

References:

Branfireun, B. A., N. T. Roulet, C. A. Kelly, and J. W. M. Rudd. 1999. In situ sulphate stimulation of mercury methylation in a boreal peatland: Toward a link between acid rain and methylmercury contamination in remote environments. Global Biogeochemical Cycles **13**:743-750.

Driscoll, C. T., Y.-J. Han, C. Y. Chen, D. C. Evers, K. F. Lambert, T. M. Holsen, N. C. Kamman, and R. K. Munson. 2007. Mercury contamination in forest and freshwater ecosystems in the Northeastern United States. BioScience **57**:17-28.

Driscoll, C. T., G. B. Lawrence, A. J. Bulger, T. J. Butler, C. S. Cronan, C. Eagar, K. F. Lambert, G. E. Likens, J. L. Stoddard, and K. C. Weathers. 2001. Acidic deposition in the northeastern United States: Sources and inputs, ecosystem effects, and management strategies. BioScience **51**:180-198.

Jeremiason, J. D., D. R. Engstrom, E. B. Swain, E. A. Nater, B. M. Johnson, J. E. Almendinger, B. A. Monson, and R. K. Kolka. 2006. Sulfate addition increases methylmercury production in an experimental wetland. Environmental Science and Technology **40**:3800-3806.

St. Clair, S. B., J. E. Carlson, and J. P. Lynch. 2005. Evidence for oxidative stress in sugar maple stands growing on acidic, nutrient imbalanced forest soils. Oecologia **145**:258-269.

Warby, R. A. F., C. E. Johnson, and C. T. Driscoll. (in press). Continuing acidification of organic soils across the northeastern USA: 1984 - 2001. Soil Science Society of America Journal.

Dr. Paul J. Hanson

The Risk and Exposure Assessment (REA) … represents a good beginning, but many sections are not yet complete or only partially complete making it difficult to judge the full intent or appropriateness of the document.

Specific comments and suggested edits:

Front matter:

Page xii: Should the definition of ecological dose be limited to toxicants that inhibit *microbe-mediated ecological processes*? I would think that the term should apply to all biological organisms. It may be that it is a term that dominates microbial studies.

Top of Page xiii: The most common example of an ecosystem benefit would probably be an increase in productivity. Why isn't this an example?

Page xiii: The difference between elasticity and sensitivity (page xv) isn't clear. Is there a reference for the use of the term elasticity that might be placed here?

Page xv: The precipitation range for semi-arid regions should probably be 250 to 500 mm. It certainly shouldn't be the same as the definition for arid systems.

Chapter 1

Page 1-3 line 8: I recommend the following wording change "…both ambient air and surface deposited species of NOx and SOx…

Page 1-10 line 23: N_2O is nitrous oxide not nitrogen dioxide.

Page 1-12 line 9: For parallel structure I would add the spelled out version of sulfur dioxide.

Page 1-13 line 5: Remove the semicolon.

Page 1-13 line 13: I would change "direct effects" to 'direct adverse effects'. There is some evidence for localized N uptake.

Page 1-14 line 11: The text must be changed to "..detail on how acidification affects sensitive ecosystems….." Don't leave the reader with the impression that acidification is having effects on all ecosystems.

Page 1-14 line 17: I don't understand "re-acidification". This concept needs to be further developed.

Page 1-14 line 31: Add 'productivity' to the list of changes driven by N deposition.

Page 1-15 line 14: This threshold for N saturation is only relevant for some, but not all eastern forests. Those levels of N deposition would be easily assimilated by much of the upland oak forests throughout the eastern United States growing on deeps soils with ample base saturation. Page 3-186 of the ISA states "there is currently no published national assessment of empirical critical loads for N in the U.S……" Table 3-25 is a nice summary of what is known. The REA needs to reflect the limited amount of data available for developing quantifiable thresholds, and appropriately characterize those ecosystems for which it might appropriately be applied. Avoid inappropriate extrapolations. Lines 14 to 19 on page 4-16 of the ISA include good statements that should be used within the REA to qualify the nature of ecosystem sensitivity.

Page 1-15 lines 21 and 22: Similarly, the wording may need to be changed here to suggest if these levels represent an appropriate threshold for all or just some sensitive grasslands.

Page 1-15 line 30: Add a reference for the sentence ending on this line.

Figure 1.3-3 still doesn't show the productivity enhancement effect of N, which is clearly a dominant process in the N cycle.

Bottom of page 1-17: Should some mention be made at this point to N standards for water pollution? It isn't necessary, but might be a useful connection. Section 3.3.6 of the ISA and the associated annex might be cited.

Figure 1.4-1 doesn't adequately capture the integrated nature of NOx and SOx pollution that is being attempted in this document. Are they truly intertwined all along this process or do they only come together after deposition takes place?

Chapter 2

In Table 2.1-1 ANC should be defined, productivity should be added as an ecosystem service where appropriate, and N leaching might be added as an indicator of terrestrial nutrient enrichment. The characteristics of sensitivity for terrestrial nutrient enrichment might be updated.

Sections of Table 2.3-1 still need to be filled in.

Chapter 3

Page 3-4 lines 6 and 7: Check the wording. 30 to 70% of the animal wastes can't be emitted as NH_3. Should it read 30 to 70% of the N losses from animal wastes?

Page 3-5 lines 10 to 15: Are there not any natural sulfur emissions in and around Yellowstone or in other hot springs areas of the country? I realize they may be inconsequential….

Pages 3-10 to 3-11: Question: Have all of the emissions reductions resulting from past clean air legislation been realized? Will new standards for ozone impact the likely deposition rates for N in the future? Should such a discussion be included someplace in this document?

Chapter 3 doesn't include much on temporal changes in deference to the maps of recent conditions. Those are fine, but I think some discussion of where we have been and where we are going should be included in the REA. Figures 2-59 or 2-103 from the ISA might be considered.

Page 3-12: Super and subscripts for charge are missing for SO_4^{-2}, NO_3^-, and NH_4^+.

Question: Do the QMAC estimates of deposition to terrestrial systems include foliar uptake of NO or NO_2 near urban areas or along major roadways where air concentrations are high enough to drive this pathway?

Table 3.2-2 is not well defined. It needs references.

I was disappointed by how much information was still not added to the document (especially in this Chapter). We are being asked to provide comment on a document that has a lot missing from it.

Captions to Figure 3.2-12, -13, and -14 need to be reworded to "…nitrogen deposition by <u>source and</u> quarter…. The most interesting data in many cases has to do with the source of N forms rather than the season of the year.

Figures 3.2-22, -23, -24, and -25 lack units (presumably kg/ha as for N).

Captions to Figure 3.2-27 and -28 need to be reworded to "…sulfur deposition by <u>source and</u> quarter….

Page 3-54: Again…we don't have much to review yet.

Page 3-54 line 14: Reword as…."Public welfare effects associated with direct exposure to NOx and SOx do not occur for current ambient concentrations."

Page 3-54 line 23: The discussion on pages 1-10 and 1-11 seems to disagree with this statement. The REA has already been defined to deal with total reactive N.

The discussion of Data and Tools (Section 3.2.2.2) should probably be presented earlier within Chapter 3 (perhaps around page 3-17).

Figures 3.2-35 and -36: Adding pixels for actual forest cover within these maps would be useful to better reflect the actual extent of the forest types. The coastal sage map (Figure 3.2-37) appears to be drawn this way.

The text for Figures 3.2-39, -40, -41, -42, -43, and -44 is way too small and the figure captions are inadequate. Please revise.

Page 3-70: At this point of the discussion it occurred to me that a case study for an area dominated by NH3 deposition (i.e., Iowa, Minnesota, Illinois) should probably be added to the REA. It wouldn't necessarily show adverse effects, but it would complete the picture of total reactive N deposition across the US. Page 3-103 of the ISA provides some rationale for not including such a case study since agricultural areas are overwhelmed by fertilizer additions. However, limited natural areas (forests, prairies) are embedded within areas dominated by agriculture.

<u>Chapter 4</u>: Incomplete and not reviewed.

<u>Chapter 5</u>: Incomplete and not reviewed.

<u>Chapter 6</u>

Figure 6.1-4: SRB should be defined in the figure caption.

Page 6-16 line 13: Remove the word "often". Fungi should probably also be recognized as an important contributor to decomposition.

Increased nitrogen does affect the N content of green leaves. Is this pattern well established for litterfall as well? Does the pattern differ by vegetation type (i.e., trees, grasses, crops…)?

Page 6-17: Some of this material might be better left to the ISA document. Lines 24 to 28 are not needed here.

Page 6-19: How the contribution of atmospheric deposition to upland watersheds actually finds its way into waterways isn't clear. The 'filtering' effect of upland vegetation will vary tremendously from location-to-location. This concept needs to be made clear in the document.

Chapter 6 seems to lack summary conclusions:

What terrestrial systems are at risk? What percent of US land area?
What aquatic systems are at risk? What percent of US freshwater area?

Chapter 7

Page 7-5 line 4: Replace "ecosystem health" with another term. It isn't and perhaps can't be defined.

Page 7-5. Line 7: Change to "this is hypothesized to change…." Or provide the references that show proof.

Page 7-5 line 21: Spell out CSS.

Page 7-7 Table 7.1-1: Replace "tree health" with a more meaningful term or terms such as rate of growth, survival….

Page 7-8: The deposition levels proposed are appropriate for 'sensitive' ecosystem, but not all ecosystems. A concept of one size does not fit all will need to be worked into the conclusions of the REA. While protection of sensitive systems may be a justification for a new standard and level, it shouldn't be interpreted as having the same effect on all areas of the US. That is, lowering inputs to areas currently unaffected will not help them. Pages 3-78 and 3-79 of the ISA include text that might be useful in clarifying this point.

Chapter 7 ends abruptly and seems incomplete.

Chapter 8

Page 8-1 lines 14 and 15: The concept of achieving a standard based on a "uniform level of ecosystem protection" seems at odds with the REA document. The REA clearly states that the impacts of N and S deposition are localized throughout the US and subject to the correct combinations of deposition and susceptibility of the target terrestrial and aquatic ecosystems.

Page 8-4 Figure 8.1-1: An air quality based estimate of total N and S form deposition may not be sufficient information to judge impacts on acid neutralizing capacity. A data layer on extant edaphic conditions is needed. The deposition metric should probably also be enhanced to allow for the estimation of biological immobilization (i.e., plant and microbial uptake of some fraction of the total deposition).

Page 8-5 lines 1 to 3: Plant uptake needs to be included in this list.

Page 8-28 lines 2 to 9: I don't agree with the assumption that annual accumulation of N inputs into wood increment can be ignored in this analysis. Except for low or no productivity ecosystems this is a significant sink for atmospheric N deposition that must be included in the calculation of N available for other soil interactions.

Section 8. 4 is incomplete. More material is needed.

Attachment 3

Page 1 line 16: The phrase "a host of biogeochemical processes" is too vague. Please expand this concept.

Attachment 4

Pages 2 and 3: Table 1.1-1 is not filled in. The term forest health should be replaced.

Pages 12 and 13: Table 1.2-2 is not complete.

Page 31: The authors conclude that the simple mass balance method would be used in the REA. What caused the authors to exclude the dynamic model method? Lack of input data? Lack of validation?

Page 32: All evapotranspiration does not occur at the surface of the soil profile. Did the authors mean to imply evaporation alone?

Page 42 line 17: Is the nitrogen immobilization mentioned here microbe and plant or just microbe?

Figure 3.1-2 is missing units.

Page 53: The conclusions need work. The imbalances for Ca, Mg, and Al suggested for forest soils are for localized sensitive systems. As worded, the conclusions would be taken as a broad generalization for all US ecosystems.

Attachment 5: No comment

Attachment 6

Page 6: Table 1.2-2 is missing too much information to be fully evaluated.

The studies cited and discussed for the CSS system should be closely evaluated to determine which were based on manipulative studies capable of determining cause-and-effect relationships versus those that represent correlation studies for which relationships between known variables and measured responses were assumed to be viable explanations for adverse responses.

Studies highlighted in the ISA within Tables 3-15, 3-17, 3-18, and 3-19 might have a larger presence within the REA.

Figure 5.1-1 and -2: In my opinion the change in CSS seen in Figure -1 doesn't correlate very well with the dominant deposition patterns in -2. How well does N deposition really correlate with change? How much does land use change through time get in the way of the interpretation of N deposition cause and effects in this case study?

Page 46 lines 4 versus line 15: The conclusion of "compelling evidence" on line 4 does not seem to agree with the authors conclusion about the research still underway on line 15. A word change seems in order.

Page 47 line 5: Is a modification of a valued ecosystem an ad

Dr. Rudolf Husar

These comments are addressing primarily the 1st draft REA section: Additional Effects. It also includes both general comments on the approach to the REA as well as comments on sections of the the first REA draft document.

Comments on REA Chapter 6: Additional Effect

1. *Charge: In this chapter, we have presented results from some initial qualitative analyses for additional effects including the impact of sulfur deposition on mercury methylation, the impact of nitrous oxide on climate change, and the impact of nitrogen deposition on carbon sequestration. Are these effects sufficiently addressed in light of the focus of this review on the other targeted effects in terms of available data to analyze them?*

Overall, this chapter is a good effort to illustrate additional, non-ecological risks associated with anthropogenic N and S. Indirect sulfur impacts are well captured by the problem of sulfur-induced methylation of mercury, which then causes the effects in biota. The interaction of nitrogen and carbon cycles is also properly illustrated in the section on nitrogen-induced changes in the carbon sequestration. The utility of the section on N_2O impact on climate is less obvious.

This chapter is clearly a first draft. In the introduction it would be helpful to provide more extensive rationale and criteria on the general approach for selecting these Additional Effects. The rationale should also include why some obvious additional effects on materials, visibility and soil are not being considered. See further discussion regarding these effects in the general comments on REA below.

6.1 Sulfur and Mercury Methylation

Page 6-2 Line 10: Currently, i.e. since 1995, coal combustion was, indeed, the main cause of anthropogenic mercury deposition. However, prior to the regulatory action, around 1990, the anthropogenic mercury emissions were dominated by other sources. Since deposited mercury has a long residence time in soil and biota, much of the mercury methylation occurs on the accumulated mercury from solid waste and other agricultural, medical, residential usages.

Page 6-2 Line 13: Atmospheric mercury particles cannot possibly remain in the atmosphere for more than two years. Even stratospheric particles, have atmospheric residence time of less than two years. As with the rest of ambient aerosols their atmospheric residence time is less than a week in the planetary boundary layer and less than 3-4 weeks in the mid-troposphere.

Page 6-3: Figure 6.1-1 represents the mercury cycle in the ecosystem. It has many compartments and arrows representing the mercury flow and transport/transformation processes. It is recognized that full quantification of the mercury cycle through air, water, land and biota is not possible at this time. However, it would be helpful, at least in the text, to highlight the main flows and processes that dominate the mercury cycle.

6.2 Nitrous Oxide

Page 6-10: I find little justification for this section on the climate impact due to anthropogenic N_2O. As stated in the REA, N_2O contributes only about 6% of GHGs and the man-induced sources of atmospheric N_2O are only 10% of that, i.e. .6% of the GHGs.

Page 6-12 Line 31: The statement "that nitrogen addition increased N_2O emission by 215%" could benefit from a reference, compared to what?

6.3 Carbon Sequestration

Page 6-13: This section is an appropriate illustration of the interdependence of nitrogen (sulfur?) and carbon cycles through air, land, water and biota. The meta-analysis of existing literature is an appropriate method for illustrating the interdependencies of the earth system components and how changes in one set of environmental chemicals may have intended and unintended consequences throughout the earth system.

General comments on the first draft REA

This REA focuses on ecosystem welfare effects that result from the deposition of total reactive nitrogen and sulfur." I concur with D. Johnson that exclusive focus on "negative" effects of N and S deposition is a flawed approach. It ignores the broader context and the full dynamics of eco system responses to anthropogenic N and S deposition. The beneficial effects of atmospheric S and N fertilization should also be considered. This would also require definitions and/or conventions on what's harmful and what's beneficial.

The man-induced nitrogen and sulfur deposition should be compared quantitatively to the naturally occurring N, S flows. This will allow estimating the significance of the man-induced stress, compared to the naturally occurring values. By avoiding such broader context, the risk assessment will be susceptible to criticisms of incompleteness and possibly irrelevance to actual ecological risk estimation.

Page 1-2 Line 9: "In the Act (Section 109 B 2) the purpose of the secondary NAAQS is to protect the public welfare from ***any*** known or anticipated adverse effects…" Clearly, a secondary NAAQS includes all welfare effects, not only the effects on the ecosystem. This fact is not followed through in the REA and ISA.

Page 1-2 Line 19: "Adverse public welfare effects are based on an assessment of how ecologically adverse impacts translate into adverse impacts on public welfare" In this sentence as well as throughout the REA welfare effects only include effects on ecosystem . As stated above welfare effects include damage to materials, visibility, soils, climate.

Specific comments on sections of the first draft REA

Page 1-13: Figure 1.3-2 representing the sulfur cycle should be made more quantitative. The schematic Figure on the biogeochemical cycles of sulfur is good. However, such a general Figure should be fortified by adding magnitudes to the flows represented by the arrows. The transfer rates, say over the US, can be estimated from the model runs, or based on the empirical evidence. There is ample literature on biogeochemical cycles that estimates the magnitude and importance of the various flow rates.

Page3-5 Line 4: " ….combustion of fossil fuels by electric utilities (~66%) " This percentage is inconsistent with 71% EGU contribution shown in the pie diagram, Figure 3.1-5.

Page 3-8: The section layout for 3.1.4 on Deposition starts with the total deposition maps that are obtained by combining the model and measured values. The description of the data and tools is given after that. The customary approach is first to present the input data, tools and methods and then the resulting computed values. Also, as discussed at the October CASAC meeting, a separate section on CMAQ model comparison with the observations for the key N, S species would be most desirable.

Page 3-8: Section 3.1.4.1 on Nitrogen Deposition has many useful quantitative numbers. However, the source and estimation methods for these estimates are not well documented.

Page 3-10: The useful Figure 3.1-7 on oxidized N deposition should be augmented with separate Figures for dry and wet deposition of N. The wet deposition Figure should also compare the model and measured oxidized N deposition.

Page 3-11: Ditto for reduced N deposition.

Page 3-12: Ditto for reduced S deposition.

Page 3-12: Section 3.2. could be subdivided such that the procedures for model-observation data fusion has a more extended separate sub-section.

Dr. Dale W. Johnson

As has been noted in previous reviews, I feel that this document is unbalanced with respect to the effects of nitrogen deposition. Some very simple facts need to be acknowledged and considered: 1) most terrestrial ecosystems in the USA are nitrogen-deficient; therefore 2) increased inputs of N are likely to cause growth increases; 3) growth increases will almost certainly result in increases in carbon (C) sequestration, which in turn may have inadvertent benefits for the CO_2 / climate problem. This is not to diminish any statements about the negative effects of N deposition, it is simply to add balance to this document. As scientific reviewers, we have the responsibility to treat this and all other subjects in a completely objective manner.

Below are some specific comments, some editorial in nature, some technical in nature, and some where I see this lack of objectivity and balance. Following that I will address the specific questions assigned to me.

Specific comments:

p. 1-13, lines 4-7: This is a balanced statement – the review of effects should really flow from this approach, considering both increased productivity (which may be beneficial in some cases, detrimental in others) and increased soil acidity and eutrophication.

p. 1-15, line 14: 5.6 to 10 kg ha^{-1} yr^{-1}? Can you really narrow this down to one decimal point?

p. 1-15, line 17: should include "and carbon sequestion" after "carbon cycling".

0. 1-17, line 13: From a soils point of view, the effects of NOx really cannot be readily distinguished from the effects due to total reactive nitrogen – both are transformed in the soil rather extensively.

p. 2-5, line 25: should add "timber production and carbon sequestration" after "water". I note that timber production is mentioned page 2-7, lines 19-20, but only in the context of how soil acidification might negatively affect it. Soil acidification may well negatively affect timber production, and the latter statement should stay as it is, but increased N deposition will probably also increase timber production and this needs to be acknowledged.

p. 2-9, line 27: should add "timber production and carbon sequestration" after "water quality"

Attachment 1, p. 3: I see Carbon Sequestration is listed as a potential section 6.3 – this is a good thing. Looking forward to seeing it.

Attachment 3, p. 1, lines 17-26: There needs to be a discussion of the effects of mineral acid anions on soil solution (what Reuss calls intensity effects, which can happen very quickly) in addition to the discussion of how they affect soils (capacity effects, which take a long time to occur). Reuss points out in his 1983 paper (Reuss, 1983) and in our small book (Reuss and

Johnson, 1985), both of which are cited later in the Terrestrial Case Study, that Al^{3+} increases to the 3/2 power of Ca^{2+}, for example, as total mineral acid anion (e.g., nitrate and sulfate) concentrations increase, and this happens even if there is NO CHANGE IN THE SOIL AT ALL. Thus, if the soil is already acid, the introduction of mineral acid anions will cause the immediate mobilization of Al and acidification of soil solutions and probably surface waters long before any change in the soil takes place. Conversely, if the mineral acid anion concentrations are reduced, one should see a very rapid recovery. In short, the soil solution can change very quickly and almost independently of the soil, and this has major implications for the effects of N and S deposition on aquatic ecosystems.

Attachment 4, p. 1. lines 16-29: Same exact comment above applies here and in this case, our small book is cited as a source but only part of the story (the soil part, not the soil solution part) is reviewed. This is an important point – please include it in the next draft.

Attachment 4, p. 5, lines 10-17: Same comment as above here. It is not necessarily true that "inorganic Al does not become mobilized until after soil Ca is depleted" if the soil is already acidic, as many unpolluted soils indeed are.

Question 4 Response: The revisions have improved the characterization of adverse ecological effects, but I see no real consideration of the potential positive effects of N deposition as yet. Timber production is mentioned, but only in a negative context and I see little or nothing on C sequestration. The one pager for section 5 refers to case studies and gives no indication that this approach will be changed.

Question 4b: I see no discussion on effects on carbon budgeting as yet – have I missed something? I do see Carbon Sequestration is listed as a potential section 6.3 – this is a good thing. Looking forward to seeing it.

Chapters 7 and 8 (Sent 19 Sept 2008)

Chapter 7: We are asked if the approach is technically sound to consider "ecologically adverse effects". For the most part, yes, it is. However, once again, I note that this section focuses entirely on negative effects of N. This document should also consider cases where increased production could be a positive effect – such as on timber production and C sequestration.

p. 7-1, lines 21-27: This is a real mouthful. Can it be simplified and broken into at least two sentences?

p. 7-3, line 12: Why "inorganic and mineral acids"? They are basically the same thing.

Section 7.1.4:

Chapter 8: Many questions are posed to the panel here and I will not repeat them. I had problems with the conceptual framework for the calculations, as noted below.

Section 8.3.3: I had a very difficult time following this section and do feel like I ought to be able to. It would help a great deal if units could be specified in the various equations and the assumptions were clearly spelled out in the beginning. For example, is it assumed that base cation concentrations will remain at pre-industrial levels? The equations would suggest so, as would the statement on p. 8-29, lines 13-15. I cannot really agree with the assumption state here that "pre-industrial base cation concentrations effectively set the long-term capacity of the catchment to neutralize acidic deposition because it represents the only source of base cation input that is sustainable over the long-term". For one thing, soils in humid regions always naturally acidify and therefore there is no long-term steady state base cation flux until soils become extremely acidic– it is always slowly decreasing. I also think that the implicit assumption here that base cation concentrations in streams will not increases over the long-term in response to acidic inputs is flawed – some soils have a very large exchangeable base cation pool and could buffer such inputs for a much longer term than the typical attention span of scientists and policy makers, let alone the public.

p. 8-29, lines 18-19: This statement makes no sense. At steady state, the leaching rate of base cations is, by definition, equal to weathering inputs, not "at lesser or greater rates".

Dr. Donna Kenski

Overarching concerns: EPA staff have clearly made a great effort in pulling this document (and its companion ISA) together, and should be commended for the high-quality (and quantity) produced thus far. Nevertheless its current incomplete status and the pressing schedule make it hard to see how all the remaining tasks can be completed, and reviewed, in sufficient time to meet the predetermined deadlines. Staff and contractors to EPA need to be realistic in judging how much can really be done in the remaining months. If it is time to set priorities, perhaps that is something that can be discussed at this meeting.

The CAA requirement that the secondary standard be in the form of a concentration standard is going to require us to tolerate a much higher level of uncertainty than usual in the standard setting process, because of the need to employ multiple models to characterize the concentration-deposition-ecological effect-ecological indicator linkage. Consequently, the REA in general needs to be much more comprehensive and transparent in describing the levels of uncertainty encountered at each of these steps and their impacts on overall uncertainty. For example, so much hinges on the CMAQ estimates of deposition, and yet there is little information given in the REA or the ISA on CMAQ performance. It seems from the ISA that CMAQ has really only been evaluated in terms of its annual estimates of aerosol deposition, and those are accurate to within a factor of 2. No CMAQ performance evaluation is given for deposition to specific locations, particularly locations that share characteristics of the sensitive areas focused on in this analysis. Likewise, none is given for measurements with a shorter time frame than annually. A clear-eyed discussion of these uncertainties for CMAQ and for the other models used to support the REA (MAGIC, ASTRAP, Sparrow) is a critical component that needs to be incorporated.

Charge Questions: Scope of the Review

1. *Is the review appropriately focused in terms of the targeted effect variables and in terms of characterizing the important atmospheric and ecologic variables that influence deposition and ultimately the ecologic impacts of nitrogen and sulfur? Does the Panel have any further suggested refinements at this time?*

Generally, the review seemed to focus on appropriate variables, although as noted above, the complete scope may be too broad to accomplish before the court's deadline. The policy-relevant questions posed in Sec. 1.4 weren't actually addressed directly (perhaps it's still too soon, given the incomplete case studies) but I did note that questions 3 and 4 of that list (i.e., to what extent do receptor surfaces influence dry deposition, and can effects of NOx be distinguished from effects due to total reactive nitrogen) did not seem to be discussed or addressed by any of the case studies in Attachments 3-6, although Chap. 3 did present a nice graphical characterization of the areas and their relative proportions of NOx vs total and other forms of N nitrogen. However, most of that was modeled data and little comparison to measured values was presented for comparison. Perhaps more of that is coming in the second draft, since there were lots of missing sections to Chap. 3.

Similarly, the list of issues on p. 1-20 should include evaluating the impacts of atmospheric deposition relative to other paths (nitrogen runoff from agricultural lands, for example). This might be what is meant by the last bullet, but it wasn't clear; perhaps it could be made more explicit.

Air Quality Analyses (Chapter 3)

1. *To what extent are the air quality characterizations and analyses presented in Chapter 3 technically sound, clearly communicated, appropriately characterized and relevant to the review of the NAAQS?*

The analyses in Chap. 3 could more accurately be described as modeled estimates of air quality, rather than air quality characterizations, which I think of as based on measured data. While the graphs were useful and logically presented, there was very little measured data given for comparison, so it is not possible to judge their 'soundness'. Combined with the lack of CMAQ validation discussion (mentioned above), it becomes more important to see these in the context of measured data as well. But this may be premature if the next draft is meant to include such comparisons.

I would have liked more discussion of the monthly patterns of deposition shown, for example, in Figs 3.2-14 through 3.2-19. Clearly wet deposition is driven largely by precipitation, but what drives the other components of deposition? Are these emission patterns or meteorological patterns or biological activity patterns? Some discussion of the importance of these various temporal scales for the ecological effects modeled is probably appropriate as well. A minor complaint on the communication of the results: the color scheme for Figures like 3.2-6 etc. is not intuitive. The scheme used was almost a rainbow-like scale, which is easy to interpret and would have been fine, but instead green was sandwiched between yellow and orange, breaking the natural progression of colors (red -> orange -> yellow) that most of us have internalized and making it harder to visually establish a continuous gradient of concentration changes.

2. *Section 3.2.1 describes an approach for evaluating the spatial and temporal patterns for N and S deposition and associated ambient concentrations in the case study locations. This document includes the analysis for the Adirondacks case study. Does the Panel agree with this approach and should it be applied to the other case study areas?*

It was a sound approach and a useful exercise that may give as clear a picture of deposition as we're likely to get. With the additions/changes noted above, I would welcome this analysis for the other areas.

3. *Section 3.2.2 describes the relative contributions of ambient emissions of nitrogen and ammonia to nitrogen deposition for the case study areas. To what extent is the approach taken technically sound, clearly communicated, and appropriately characterized?*

This was an interesting exercise that was very useful in establishing the relative importance of NOx and NH3 emissions to overall N deposition and the relative responsiveness of deposition to changes in emissions. It was very helpful to establish this kind of personal internal calibration in a strong visual way. It is also a critical comparison to establish in making a strong case for the ability of a NOx-concentration based standard to be sufficiently protective, despite our understanding that impacts are really driven by total nitrogen. Given the lack of spatial and temporal variability in the contributions of NOx to oxidized N and NH3 to reduced N, those maps probably don't need to be shown, just described instead. The results look perfectly logical and convincing, although I resist accepting these results completely until seeing further documentation of CMAQ's performance. The REA notes that the RSM has been validated for PM and O3, but it's not clear whether that validation translates to deposition parameters as well. Presumably the missing section 3.2.2.5 on uncertainty will address some of these issues. Again, as above, the color scale on all these plots is counterintuitive. Using the color red to depict no impact and green for 100% impact is contrary to general mapping conventions and at least in my case, caused me to continually misinterpret the plots.

Case Study Analyses (Attachments 2-6)

6. *Att. 2: Are the national geospatial datasets chosen adequate to identify sensitive areas? Are there other data sets that have not been identified by this analysis that we should consider? Does the panel agree with this approach or can they suggest alternatives?*

I have no knowledge of other datasets that could be useful to this effort. Some of the data were quite old (1971 for the range of red spruce) and caused me to wonder whether the range could have changed significantly in the intervening 37 years. Perhaps the authors could comment on the issues that might be affected by such old observations. Similarly, the dataset on acidophytic lichens was clearly not complete, or at least spatially representative, and impacts the results. One can't protect lichens that haven't been identified as sensitive, and the current map, which shows clusters of lichens within some states but none in neighboring states, strongly implies that some species have not been identified in those neighboring states.

7. Att. 3: re MAGIC model to evaluate ANC levels in selected streams and lakes in Adirondacks and Shenandoahs. To what extent is the approach taken technically sound, clearly communicated, and appropriately characterized?

The selection of MAGIC is appropriate, but this section was poorly written and extremely confusing, especially the modeling approach and description of MAGIC and ASTRAP. I think the dates given for scaling the historic data to deposition are wrong in several places, but the text was too convoluted to tell for sure. Also it would have been nice to see a map or at least a better description of how the ASTRAP sites are connected to the MAGIC sites. The classes and descriptions of ANC limits kept changing within the text and figures and should be made consistent. The discussions of critical load frequently mixed up the concepts of greater than-less than and above-below, adding to the confusion. It was very difficult to wade through the errors and try to make sense of what was really being accomplished here. The approach may be sound, but can't be assessed on the basis of what was presented. It was certainly not communicated or characterized in a satisfactory way.

8. *Att. 4: use of SMB model to evaluate current deposition on forest soil ANC for sugar maple in Kane Experimental Forest and red spruce in Hubbard Brook Experimental Forest. To what extent is the approach taken technically sound, clearly communicated, and appropriately characterized?*

The selection of the study area was reasonable and the description of the method was clear. Without results it's not possible to say too much more. The Table 3.1-2 had some numbers that need to be explained further, however. The range of critical N loads vary by a factor of 10 over the 3 study periods shown, and each of the methods gives very different results. Why the big difference? The text mentions biomass changes; is this the sole reason? Is this magnitude of change in biomass typical? How comparable are the methods? The text in Section 1.1.1 was unnecessarily repetitive and could be tightened up; no need to quote the ISA at such length.

9. *Att. 5: Aquatic nutrient enrichment—evaluate how changes in N deposition affect the eutrophication index in two estuaries: Chesapeake Bay and Pamlico Sound. To what extent is the approach taken technically sound, clearly communicated, and appropriately characterized?*
10. *Att. 6: Terrestrial nutrient enrichment—evaluate effects of N deposition on CSS community in California and mixed conifer forests in San Bernardino and Sierra Nevada Mountains. To what extent is the approach taken technically sound, clearly communicated, and appropriately characterized?*

Additional Effects (Chap. 6)

1. *Impacts of S deposition on Hg methylation, impacts of NO on climate change, and impact of N deposition on C sequestration. Are these effects sufficiently addressed in light of the focus of this review on the other targeted effects and in terms of the data available to analyze them?*

This was an adequate review of these effects. The focus appropriately belongs on acidification and enrichment effects.

Synthesis and Integration of Case Study Results into the Standard Setting Process (Chap 7)

1. *Purpose is to summarize the case study results and characterize the relationship between levels of an ecological indicator and the associated degree of ecologically adverse effects. To what extent is the approach taken technically sound, clearly communicated, and appropriately characterized? Does the Panel have suggested refinements?*

I'm still uncertain about exactly how the case studies ultimately are used. Are they merely for scientific support and justification, or do the model results from them get incorporated eventually into a quantitative relationship that can be plugged into the framework presented in Chapter 8? I think the confusion stems from the incomplete nature of this chapter. When it is fleshed out in the next version with real data, the application of the case study results should be obvious.

Considerations in the Structure of the NOx/Sox Secondary Standard (Chap 8)

1. *Is the suggested overall structural framework for a secondary standard technically sound and logically presented?*

The recently added Chapter 8 was very lucid and well written, presenting a helpful blueprint for how to move from ambient concentrations to possible secondary standard. I found no technical or logical fault in it.

2. *Is the description and development of the deposition transformation function (Fig 8.1.1) between air quality indicators and the deposition metric clear and technically sound?*

3. *Is the description and development of the ecological effect function (Fig. 8.1.1) between the deposition metric and the ecological indicator clear and technically sound?*

4. *Does the discussion adequately capture the potential use of categorical variables versus continuous variables when accounting for the variability of atmospheric, landscape, and ecological factors?*

These were all fine.

5. *Are there any key elements missing from the framework?*

There needs to be a more complete discussion of how a standard can accommodate geographic variation; i.e., how to equitably control NOx and Sox to adequately protect sensitive areas without overcontrolling in areas that are far less sensitive to acid deposition. Section 8.3.1 made reference to varying factors within the transformation function by location, and the fact that deposition loads will vary by location, but it was not clear from the text how this translates to variation in the standard. It seems that the standard must account for different concentrations in different regions, but how that is specified is still pretty fuzzy.

Another big piece that is missing is the discussion of CMAQ (or other model) uncertainties. It was interesting that this is the first place in the REA (Sec. 8.3.2.1.2) where CMAQ performance evaluation (or the lack thereof) is mentioned. Given the very heavy reliance of the REA analyses on CMAQ (a necessary reliance, admittedly), it is critical to have its performance characterized as thoroughly as possible so that the Administrator and others can assess a proposed standard's potential for success.

Section 8.3.2.2.2 leaves aggregation methodology as an open research issue. Does this mean it will be resolved in the next draft by work the staff is doing? If not what are the implications of leaving this issue unresolved?

6. *Does the framework need to be expanded or revised to accommodate the appropriate consideration of ecological indicators besides ANC when developing a secondary NAAQS for NOx and Sox*

Isn't that the point of the case studies? It seems premature to answer this question until those results are complete. ANC seems adequate for acidification effects, but the other case studies weren't really far enough along to evaluate. The chapter should include some discussion of how the standard might account for multiple indicators.

7. *Does the panel have any further suggested refinements at this time?*

Section 8.3.2.4 needs additional attention. It mentions possible work on MCIP and CMAQ to incorporate measurements, which would be great and I highly encourage. But is it really possible to produce results and incorporate them into the next draft?

Section 8.3.3 on the deposition-indicator links could benefit from a discussion of how influential the various parameters are on the outcome, at least in a qualitative sense. It would help the reader (and presumably the Administrator) evaluate which are the critical parameters controlling the relationship. There also needs to be some discussion or graphical analysis of the geographic sphere of influence of NOx/SOx concentrations on each sensitive area. For example, if the indicator were set at an ANC of 50 ueq/L in the Adirondacks, how much would NOx and Sox need to be reduced to reach that ANC, and over what geographic area? Somewhere the document needs to lay out that visual frame of reference for the geography of influence on the sensitive areas. We know that deposition is some function of distance, and that high concentrations close to a sensitive receptor will be more influential than the same concentrations far way; how far away is far enough to have negligible impact?

Specific comments, typos, etc.:

p. 3-3, Fig 3.1.2 (also Fig 3.1-5) Avoid use of pie charts, as they make it difficult to make quantitative comparisons. Bar charts are almost always preferable. The use of 3-D unnecessarily complicates these figures and also makes it more difficult to visually compare the slices of the pies.
p. 3-4, lines 12-13: Fig. 3.1-4 actually shows facilities, not annual emissions by state.
p. 3-6, lines 14-16: Is the NEI fire inventory error corrected here? If not, a statement about the correct magnitude of fire emissions should be added.
p. 3-8, line 9: Figure 3.1-6 shows total N deposition in Ohio and Pennsylvania of >20 kg/ha/yr, definitely much more than the 9.2-9.6 kg/ha/yr cited here. Which is correct? This and the following figs 3.1-7,8,and 9 are nice but the colors are difficult to distinguish in the printed version.
p. 3-50, line 1: ARD -> ADR
p. 3-57, line 10: remove question mark
p. 6-7 caption to Fig. 6.1-3: watershed should be plural
p. 6-18, line 6: Not clear what 'further stabilizing soil carbon compounds' actually means. Do they then have longer lifetimes?
p. 6-23, line 12: Onondagal ->Onondaga
p. 7-1, line 28: area should be plural
p. 8-14: Equation numbers don't follow the text

Attachment 2, p. 2, line 14: remove 'is'
Attachment 2, p. 3, line 19: is this really 51 inches, or should it be cm, as 4.1.11 says about this same dataset?
Attachment 2, p. 7, line 17: remove 'Sulfur Containing'
Attachment 2, p. 7, line 26: remove 'Nitrogen Containing Chemical Species'
Attachment 2, p. 8, line 9: remove 'deposited'

Attachment 2, p. 11, line 8: remove 'Nitrogen Containing Chemical Species'
Attachment 2, p. 11, line 18: Containing -> Including
Attachment 2, p. 16, line 4: remove meter

Attachment 3: fix the subscripts and superscripts throughout. Too many grammar errors and typos to enumerate here – this whole section needs careful editing.
Attachment 3, p. 6, line 24: deposition is more like 17 and 13 kg/ha according to the Figure 3.1-1, not 15 and 10.
Attachment 3, p. 8, line 4: deposition is more like 18 and 11 kg/ha according to Fig. 3.2-1. This makes me distrust the % declines in these species, here and on p. 6, but I didn't recalculate them.
Attachment 3, p. 12, Fig. caption: 12 streams are shown, not 13. Lots of other typos in this caption.
Attachment 3, p. 16, line 26: >50 should be <50
Attachment 3, p. 17, lines 6-8: Fig. 4.1-2 doesn't imply that biota are not often harmed below and ANC of 100, only that the harm is less severe than for lower ANC
Attachment 3, p. 17, line 12: It doesn't make sense to say that an ANC of 0 protects surface waters from becoming acidic. waterbody
Attachment 3, p. 17, line 17: change 'deposition – critical load' to 'deposition less than critical load'

Attachment 4, p. 1, line 6: should be 'sulfur loads *to* and effects *on* a chosen…'
Attachment 4, p. 10, line 4: on -> at
Attachment 4, p. 11, line 16: not clear, reword
Attachment 4, p. 11, lines 21-22: not clear, reword
Attachment 4, p. 23, line 22: HBEF
Attachment 4, p. 25, line 8: litterfall
Attachment 4, p. 33, line 17: put weathering on its own line
Attachment 4, p. 38, line 13: Arrhenius
Attachment 4, p. 48 and 49: It would be helpful for these figures to include the HBEF as well; its very hard to place it accurately given the map on p. 21

Dr. Naresh Kumar

COMMENTS ON SECTION 6.1. SULFUR AND MERCURY METHYLATION

GENERAL COMMENTS

The section begins with a segment on the chemistry and physics of mercury atmospheric transport, fate, and deposition, followed by a more detailed segment discussing the chemical determinants of mercury methylation.

The two segments of the section differ greatly in their accuracy, completeness, and understanding of mercury chemistry and environmental behavior. While the second part, on mercury methylation specifically, is generally complete and accurate, the first, background, segment has a number of factual errors, misinterpretations, and erroneous conclusions in it.

As two examples, the first segment presents an erroneous picture of the behavior of elemental mercury (or Hg^0) in the environment, and of "methylmercury" (dimethylmercuric salts) in organisms. In the first case, Hg^0 is presented as being "reduced" in surface ecosystems to become methylmercury; in reality, Hg^0 is the reduced form, and plays no part in methylation, which occurs through bacterial action on the oxidized form, divalent mercury (or Hg^{+2} in the text). In the second case, methylmercury is openly stated to be "lipophilic," or preferentially attached to fatty tissues in fauna, when in fact methylmercury is lipophobic and associated with protein sulfhydryl groups, in muscle tissue.

The entire section need to be thoroughly reviewed and rewritten from the beginning to more accurately reflect our basic understanding of mercury chemistry, transport, and fate in the environment. Specific comments on the text follow.

SPECIFIC COMMENTS

- To date, there has been *no* unequivocal demonstration of sulfate limitation in US or global waterways such that natural sulfate addition or subtraction alone has produced a change in methylation rates or mercury in fish. Such demonstrations have occurred only in experimental manipulations of microcosm ecosystems. Since fish take up only a fraction of the methylmercury in the water column and biota of lower trophic levels, there is always an excess of MeHg in studied water bodies. And downtrends in sulfate addition have always been matched by downtrends in divalent mercury deposition, so that it is not possible to separate sulfate availability from divalent mercury burden.

- It is mentioned that *"Mercury concentrations have increased approximately 2 to 5 times since the onset of the industrial revolution and appear in even the most remote locations on the Earth (Munthe et al., 2007; U.S. EPA, 2006)."* The accepted global average ratio of atmospheric mercury mass now compared to the period prior to the Industrial Revolution is in the range of 2 to 3, not 2 to 5. Ratios higher than 3 can be found in local, single-instance measurements of concentrations in, e.g., an ice core, but these are characteristic of individual locations and not the global balance of mercury.

- It is stated in the text that *"In the United States, the primary source of mercury to ecosystems is atmospheric deposition due to coal combustion (e.g., coal-fired electric utilities). Other sources include municipal waste combustion, medical waste incineration, chlor-alkali plants, and industrial boilers."* This sentence appears to propagate the common misconception that mercury emissions (anywhere) are proportioned exactly the same as mercury deposition (anywhere else). This is obviously incorrect, since both total and wet deposition of mercury at any location on earth (or in the United States) is made up of contributions from hundreds of sources at widely varying distances upwind, and is not linearly proportional to the fraction of total emissions each source, or source type, makes up. Therefore, it should be noted that a significant amount of mercury depositing within the United States originates in other countries, primarily mainland Asia. The sources of mercury depositing to U.S. ecosystems varies widely, both geographically and by source, depending on the proximity of U.S. sources and the precipitation climatology of the setting.

- It is mentioned that, *"Depending on the particulate association and oxidation state, atmospheric mercury particles can remain suspended in the atmosphere for more than 2 years (Evers et al., 2007; U.S. EPA, 2006)*." This sentence should be rewritten or *deleted.* There is no relation between "particulate association" (an unexplained term) and oxidation state for mercury; most of the mercury bound to particles is divalent mercury (or "Hg^{+2}" as used in the report). But the statement that such particle-bound mercury has an atmospheric lifetime of more than 2 years is not true; due to gravitational settling, coagulation, etc., Hg_p has an average lifetime in the atmosphere of several days to about two weeks, no more. And no source of mercury emissions to the atmosphere issues more than about 3% of mercury mass in the form of particle-bound mercury, in any case. It is suggested inserting something like this: *"There are three primary forms of mercury in atmospheric sources of the substance: elemental mercury, reactive gaseous mercury, and particle-bound mercury. Once emitted, the three forms behave very differently in the atmosphere and deposit over very different geographic patterns. It generally takes hundreds or thousands of miles for half of the emitted gaseous elemental mercury to deposit to ground level, while half of the reactive gaseous mercury will deposit within about 150 miles of the source. Particulate-bound mercury, generally 3 percent or less of the emitted mercury mass, deposits in intermediate patterns (M. Cohen, 2004)."*

- *The text states that, "When deposited into terrestrial and aquatic ecosystems, elemental mercury is oxidized to reactive mercury (Hg^{+2}) (Ambrose et al., 2005; U.S. EPA, 2006)."* A number of statements need to be corrected or nuanced in this passage. Any "deposition" of elemental mercury occurs by gas-phase transfer at ground level from regions of higher concentration (that is, the atmosphere) to regions of lower concentration (to plant stomata, soil pores, interstitial spaces, etc.), basically a down-gradient mass transfer. There is nearly no oxidation of elemental mercury to the divalent form occurring at the ground surface; more likely is removal of elemental mercury back to the atmosphere by revolatilization, or evasion. There may also be a net output of elemental mercury from the surface by insolation (solar radiation) producing photoreduction of divalent mercury, or demethylation and photoreduction of monomethylmercuric halides.

- The entire sentence where methylmercury is stated to be "lipophilic" needs to be corrected. First, mercury cannot be "reduced and methylated to methylmercury"; the reduced form of mercury is the insoluble elemental mercury, Hg^0; because of its insolubility in water, it is unavailable to sulfate-reducing bacteria for the methylation process. Of the "deposited mercury pool," typically half or more is (wet-deposited) divalent mercury, most of the remainder dry-deposited elemental mercury; of the amount of divalent mercury dissolving in water bodies, between 1% and 10% may be methylated and dissolved in the water column. Of this 1% to 10% (depending on the particular water chemistry; higher fractions for more anoxic waterways), perhaps 10% of that (or 0.1% to 1% of the dissolved divalent mercury) may be taken up into the food web. Second, mercury is most certainly *NOT* lipophilic, but rather ***lipophobic***: it attaches to protein-based sulfhydryl groups and resides primarily in muscle and nerve tissue ("fish flesh"). This distinction is important because it is the root of the finding that cooking fish which may be mercury-laden will in fact not decrease, but increase, the concentration of the mercury in the cooked product. Any fat that is cooked off is mercury-free, and the lower weight cooked fish remaining has the same mass of mercury as prior to cooking, but in a lower-weight portion of fish (with some fat and water mass cooked off), hence higher net mercury concentration.

- There seems to be a faulty reference in the sentence ***"****The majority of U.S. waters are sulfate-limited (Harmon et al., 2007); therefore, decreases in sulfate are likely to promote decreases in methylmercury."* Harmon et al., 2007, "Using Sulfate-Amended Sediment Slurry Batch Reactors to Evaluate Mercury Methylation," *Arch. Environ. Contam. Toxicol. 52*, 326–331 (2007); [DOI: 10.1007/s00244-006-0071-x] does not have a single word to say about sulfate-limited waterways. The term "-limited," in fact, occurs only once in the document, in the introduction, with no reference to the state of U.S. waters.

Dr. Myron J. Mitchell

General Comments (For these general comments my responses are in *italics*.)

The document and associated attachments are extensive and it is a major challenge to provide a cohesive approach to presenting this information. I recognize the difficulty and complexity of pulling all of this together. The potential for providing clearer linkages in this document to the "Integrated Science Assessment for Oxides of Nitrogen and Sulfur – Environmental Criteria" may help maintain focus and reduce the size of the document. More consistency is needed in the use of terms such as oxides of sulfur, oxides of nitrogen, sulfur oxides, nitrogen oxides, SO_x, NO_x, *etc. Also, in the document there are differences in criteria for sensitivity of various parameters such as ANC. Careful editing would help focus the document and reduce redundant information. The draft document has many noun trains such as "draft revised criteria document".*

Responses to Charge to the CASAC NO_x/SO_x Secondary Review Panel

Scope of the review (Chapters 1 and 2):

1. Chapters 1 and 2 provide the background, history, and the framework for this review, including a discussion of our focus on the four key ecological effect areas (aquatic acidification, terrestrial acidification, aquatic nutrient enrichment, terrestrial nutrient enrichment). Is this review appropriately focused in terms of the targeted effect areas and in terms of characterizing the important atmospheric and ecological variables the influence the deposition and, ultimately, the ecological impacts of nitrogen and sulfur? Does the Panel have any further suggested refinements at this time?

Chapter 1 needs a more balanced introduction with respect to the impacts of S and N gaseous constituents and deposition with greater attention earlier in each chapter on the effects of N on ecosystem structure and function including the alteration of species composition. In Chapter 2 (Section 2.4) the description of errors in the analyses needs a more rigorous approach that more clearly identifies specific issues related to sources and amount of errors. In its current form Chapter 2 provides very broad descriptions that do not clearly identify the major sources of error that are important to the current assessment. Possibly reference to subsequent chapters that more clearly delineate these errors would be helpful.

Air quality analyses (Chapter 3):

1. To what extent are the air quality characterizations and analyses presented in Chapter 3 technically sound, clearly communicated, appropriately characterized, and relevant to the review of the secondary NAAQS for NO_x and SO_x ?

The terminology and analyses associated with "policy-relevant background concentrations" is confusing. Some consideration is needed of the adequacy of the current monitoring network in *evaluating air quality especially in relationship to measurements in urban settings.*

2. Section 3.2.1 describes an approach for evaluating the spatial and temporal patterns for nitrogen and sulfur deposition and associated ambient concentrations in the case study locations. This draft document includes the analysis for the Adirondacks case study. Does the Panel agree with this approach and should it be applied to the other Case Study Areas?

This can be a helpful approach. However, within each of these regions there have been other measurements made of associated parameters including precipitation amount, atmospheric deposition, etc. It does not appear that any substantial attempts are being made in this document to compare these current results with previously published results? There is a major emphasis on providing both spatial and temporal patterns using the CMAQ modeling results. Shouldn't these modeling results be compared with other results of deposition measurements for this region? Having some comparisons with these other measurements would be helpful in evaluating the CMAQ deposition estimates. The issues and problems associated with making measurements of air pollution concentrations and converting them to deposition by various inferential procedures needs to be clearly articulated.

3. Section 3.2.2 describes the relative contributions of ambient emissions of nitrogen and ammonia to nitrogen deposition for the case study areas. To what extent is the approach taken technically sound, clearly communicated, and appropriately characterized?

Clarification is needed on the relative importance of the emission sources including the accuracy of these emission estimates, atmospheric chemical conversions and the estimates of deposition velocities in evaluating the results of these model outputs. Too much of this section describes model output with little supporting evidence of the relationships noted.

Case Study Analyses (Attachments 2-6):

These attachments are extensive and it would be most helpful if they were more targeted to the issues and needs of the "Risk and Exposure Assessment to Support the joint Review of the NO_2 and SO_2 Secondary National Ambient Air Quality Standards".

1. Attachment 2 presents a GIS analysis to define geographical areas that are sensitive to acidification and nutrient enrichment. Are the national geospatial datasets chosen adequate to identify sensitive areas? Are there other data sets that have not been identified by this analysis that we should consider? Does the Panel agree with this approach or can they suggest alternatives?

In general, the selected regions and associated data sets appear to be appropriate for this analysis.

2. Attachment 3 presents our current progress on evaluating the effect of aquatic acidification in the Adirondacks. It describes the use of the MAGIC model to evaluation ANC levels in selected lakes and streams in the Adirondacks and Shenandoahs. To what extent is the approach taken technically sound, clearly communicated, and appropriately characterized?

There is some differences in the deposition estimates being used for estimates using the MAGIC versus other approaches in the document (e.g., ASTRAP versus CMAQ). Will this result in some issues relating to congruity of analyses in the document? These analyses have a specific focus on the Grimm and MAGIC models. There have been other efforts to evaluate acid rain/biogeochemical responses in each of these sites (especially the Adirondacks) and hence it would be useful to include some of these other results to indicate either differences or support of these current modeling efforts. In these discussions different levels of ANC are suggested that differ than for other sections in the document. A more consistent approach is needed on setting the ANC limits of concern with respect to sensitivity to acidification and recovery from acidification. Possibly a separate section is needed with respect to ANC and its application to evaluating acidity and the various levels that are used. This would need to be included before the case studies section.

3. Attachment 4 presents our current progress on evaluating the effect of terrestrial acidification. It outlines a plan to use the Simple Mass Balance Model to evaluate current deposition levels on forest soil ANC for sugar maple in the Kane Experimental Forest and red spruce in the Hubbard Brook Experimental Forest. To what extent is the approach taken technically sound, clearly communicated, and appropriately characterized?

This portion of the document is highly uneven with in some cases substantial detail provided and in other places there are broad generalizations. This makes it difficult for the reader to ascertain the salient points.

4. Attachment 5 presents our current progress on evaluating the effect of aquatic nutrient enrichment. It outlines a plan to evaluate how changes in nitrogen deposition affect the eutrophication index in two estuaries: the Chesapeake Bay and Pamlico Sound. The analysis will model one stream reach (Potomac River and Neuse River) to determine the impact on the eutrophication index for the estuary. To what extend is the approach taken technically sound, clearly communicated, and appropriately characterized?

The Chesapeake Bay has been the focus of considerable efforts relating to the effects of nitrogen loading on eutrophication. There have been extensive investigations on this site and there are extensive data sets that can be used in the analyses. A major drawback of this site, however, is that it is not likely that atmospheric nitrogen loading is a major component of the total nitrogen loading. Hence, the Chesapeake Bay will not be very sensitive to changes in atmospheric nitrogen inputs. Although the information on the Neuse River is less extensive, there are similar problems with the overall importance of atmospheric N loading. In attachment 5, page 20 it is indicated that "Previous studies have estimated that wet atmospheric deposition of nitrogen (WAD-N), as deposition of dissolved inorganic nitrogen (DIN: NO^{-}_{3} , NH_3 /NH^{+}) and dissolved organic nitrogen, may contribute at least 15% of the total externally supplied or "new" nitrogen flux to the coastal waters of North Carolina".

5. Attachment 6 presents our current progress on evaluating the effect of terrestrial nutrient enrichment. It describes an approach to evaluate the effects of nitrogen deposition on the Coastal Sage Scrub community in California and in mixed conifer forests in the San Bernardino and Sierra Nevada Mountains. To what extent is the approach taken technically sound, clearly

communicated, and appropriately characterized?

The selection of the Coastal Sage Scrub community in California and in mixed conifer forests in the San Bernardino and Sierra Nevada Mountains needs some further justification versus other regions including the alpine and subalpine communities of the eastern slope of the Rocky Mountains in Colorado. These sites in California are certainly important and interesting sites, but it may be difficult to separate the effects of nitrogen deposition versus other air pollution components, especially ozone.

Additional Effects (Chapter 6):

1. In this chapter, we have presented results from some initial qualitative analyses for additional effects including the impact of sulfur deposition on mercury methylation, the impact of nitrous oxide on climate change, and the impact of nitrogen deposition on carbon sequestration. Are these effects sufficiently addressed in light of the focus of this review on the other targeted effects and in terms of the available data to analyze them?

These descriptions seem adequate, but certainly the issues go beyond those associated with carbon sequestration. Some additional discussion of the importance of mercury being found in other components of the ecosystems including song birds should be included.

The purpose of Chapter 7 is to summarize the Case Study results and characterize the relationships between levels of an ecological indicator and the associated degree of ecologically adverse effects. To what extent is this approach technically sound, clearly communicated and appropriately characterized at this point of the review? Does the Panel have any further suggested refinements at this time.

In its current form this Chapter is highly descriptive. Future revisions will need to include more specific quantitative results that show a clear relationships to the assessment.

Chapter 8 begins to explore how a secondary NAAQS might be structured to address the targeted ecological effects discussed in the risk assessment. The next draft of this document will include one or more examples of how this structure might be used to related specific levels of air quality indicators with a corresponding ecological indicator for a given location and/or scenario. To what extent is the described approach technically sound, clearly communicated and appropriately characterized at this point of the review? Specifically, we are asking:

- Is the suggested overall structural framework for a secondary standard technically sound and logically presented?
- Is the description and development of the deposition transformation function (Figure 8.11) between air quality indicators and deposition metic clear and technically sound?
- Is the description and development of the ecological effect function (Figure 8.1-1) between the deposition metric and ecological indicator clear and technically sound?
- Does the discussion adequately capture the use of categorical variables versus continuous variables when accounting for the variability of atmospheric, landscape and ecological factors?

- Are any key elements missing from the framework?
- Does the framework need to be expanded or revised to accommodate the appropriate consideration of ecological indicators besides ANC when developing a secondary NAAQS for NOx and SOx?
- Does the panel have any further suggested refinements at this time?

Providing a clear scientific linkage that is policy relevant between concentrations of SO_x and NO_x and resultant S and N deposition and subsequent translation to ecosystem level effects is a major challenge. There are good linkages between concentrations of SO_x and NO_x and deposition, but the linkage to ecosystem level effects is more complicated due to inherent variability across the United States of the sensitivity of these systems. There are major problems with respect to both having confidence in calculated deposition velocities and how to ascertain "functional forms". A clear delineation of which ecosystem types are most important with respect to specific ecological effects (e.g., acidification in the northeast, N deposition affecting biological community structure in the mountain west, etc.) is required. Such categorization will be very important in setting secondary standard(s) that will be relevant to those effects which are of greatest importance to various regions that have different relative representations of various ecosystem types.

Specific Comments in which *italics* indicate specific changes.

Page(s)	Lines(s)	Comment
xv	3-5	Why should this definition only include terrestrial ecosystems?
1-1	8	Change to "The NAAQS *have been* established for pollutants".
1-2	8	Change to "complex interactions among relevant *chemical* species of".
1-3	10	Clarify more specifically who is "we". Do you mean EPA?
1-3	19	"Chapter 2" should be in bold font.
1-4	1-5	This does not agree with provided definition on nitrogen enrichment (page xv, lines3-5)
1-5	13	Change to "*This draft*".
1-5	15	Delete "held".
1-5	24	Change to "identified the critical *components* to be".
1-5	25	Specify the title of the "Administrator" here and elsewhere in the document.

1-6	5	Change "NAAQS for NO2 *was set at* 0.053 ppm".
1-6	10	Change to "secondary NAAQS values for SO_2 under".
1-6	13	Change to "of evidence from vegetation effects ".
1-6	19	Change to "EPA was aware that SO_x has other".
1-6	25	Here and elsewhere be consistent in the use of terms SO_x and sulfur dioxides.
1-7	17	Change "completed by EPA at *this time*".
1-7	20-23	Change to "Administrator's conclusions that (1) based upon the then-current scientific understanding of acid deposition, it would be premature and unwise to prescribe any regulatory control program, and (2) when the scientific uncertainties had been *substantially* reduced through ongoing research efforts, EPA would draft and support an appropriate set of control measures".
1-8	3	Change to "*Due to* the complexities and *substantial* remaining".
1-8	20-21	Change to "long-term".
1-9	28	Be more explicit with respect to this "Federal Register notice".
1-10	7	Why "beyond full implementation of Title IV.…"?
1-10	7	Change "to presented in the *2005 NAPAP* Report".
1-10	10	Be more specific with respect to which "recent reports".
1-10-11		It would preferable to start with a clearer definition of the different forms of nitrogen oxides and then discuss some of the confusing terminology.
1-11	6	Are another group of individuals using different terminology for nitrogen oxides?
1-11	12	Change to "deposition of *reactive* nitrogen"
1-11	19	Change to "In many *regions*".
1-11	22	Change to "assessing *the impacts of nitrogen deposition*".
1-11	24-26	This is not true most research does consider separately the oxidized

		and reduced forms of nitrogen inputs especially for natural ecosystems. In many cases the total input of reduced nitrogen is less well known than that of oxidized nitrogen.
1-12	13-14	With reduction in S deposition the relative importance of N deposition in contributing to acidification has increased. This should be noted in this introductory information.
1-13	5	The statement "sometimes limiting" with respect to N is misleading. Nitrogen is often a limiting nutrient with respect to primary production in terrestrial ecosystems. Even though N may be a limiting nutrient does not necessarily imply, however, that there may not be substantial ecosystem damage. Changing N inputs can alter ecosystem structure including altering the system from a more desirable state. Such factors are especially important with respect to the evaluation of N deposition in the western U.S. Such secondary effects need to be considered in setting standards.
1-13	6	Change to "acidification, nutrient enrichment, eutrophication, and changes in species composition".
1-14	7-9	Other effects including changes in species composition and community structure need to be included for both terrestrial and aquatic systems especially associated with N inputs.
1-14	10-24	Clearly there have been substantial studies of acidic deposition effects in the eastern U.S., but some consideration is needed of effects in the western U.S. where the effects appear to be substantially different especially with respect to N deposition influences.
1-14	26	Does the reference to "high elevation lakes" refer to the western U.S.?
1-14	31	In this section the issue of changes in "community composition" is mentioned and this needs to be included earlier in the document.
1-15	1-4	This statement on alteration of aquatic communities needs to be made congruous with earlier statements on overall effects of S and N deposition.
1-15	22	Change to "*a*lpine".
1-16	5	Provide some information on what are these "other factors". Do you mean periods of relatively high microbial activity?

1-16	8	Be more explicit on the relationship (e.g., positive or negative) between "oxygen content, temperature, pH, and supply of labile organic carbon".
1-16	11	Be more explicit on what are these conditions. Do you mean for conditions the amount of wetlands and linkages to drainage waters?
1-16	Figure 1.3-3	should explicitly have a category for changes in species composition that may differ from altered biodiversity.
2-1	9-12	This listing needs to explicitly include effects on community structure and species composition.
2-1	16-18	This statement begins to raise the issue associated with different effects depending on region in the U.S.
2-1-2		The absence of case studies in the Rocky Mountain west is problematic.
2-4		This introduction can be improved with a more focused approach. There is considerable repetition in this part of the document.
2-5	4-5	Change to "data *were not sufficiently available* to perform".
2-5	18	Clarify in scaling-up clarify how this will be done with respect to sensitive areas (e.g., will this be done for all sensitive areas or will the scaling be a function of other sensitive areas with similar characteristics?
2-6	27	Change to "*need* for them".
2-6	28	A citation is needed for the "MEA" here. Later it is indicated that MEA (2005).
2-7	3	Care should be made in the use of the term "regulating" since this term has a different meaning in the context of environmental regulations such as those associated with laws formulated by the EPA. Maybe a clarifying phrase is needed to avoid any confusing recognizing that the term regulation has a different meaning in an ecological context.
2-6	14	Are there different references for the MEA? Here it is indicated: MEA, 2005b. Also see Line 17 (MEA, 2005b).
2-7		This section needs some further articulation with respect to the

		description of ecosystem services. Is this description based solely on what is provided in (U.S. EPA, 2006)? It would be helpful to provide some further references that link these statements to other informational sources.
2-7	16	Change to "interest in the risk assessment *described in the current document*".
2-7	20-22	A better description is needed that clarifies the association of NO_x deposition and eutrophication.
2-7	23-25	This statement is poorly worded.
2-7	26-31	The wording of this section needs much better clarification. It is confusing whether a linkage or several linkages are being evaluated.
2-8	Figure 2.2-2	Ecosystem Services: the ecological processes or functions having monetary or non-monetary value to *individuals or society*.
2-9	1-2	Change to "We have begun identifying the primary ecosystem service(s) affected by *either* acidification *and/or* enrichment and for major ecosystem types and components (i.e., terrestrial ecosystems, soils".
2-9	4	Change to "The impacts *affecting* various".
2-9	6-7	Change to "*These impacts on ecosystem services* will be".
2-9	10	Change to "tourism. *Effects on fisheries* (decreased".
2-9	21	Change to "tourism. Effects on fisheries".
2-9	27-28	Change to "hazard mitigation. *Methods for evaluating linkages to measurements and ecosystem* services may include".
2-10	5	Clarification is needed on what is meant by "community".
2-10	7	Change to "changes in stand density, shifts in". Densification is jargon. Change to "shifts in lichen community species composition".
2-10	14	Delete "intake".
2-10	15-16	Why distinguish between "Native Americans and Alaska native Villagers"?

2-13	2-4	Change to "*Our approach in evaluating risk assessment relies upon various analytical tools and techniques, data sources, and other forms of analyses each of which has inherent uncertainties* ".
2-13	6	Change to "affect the *level* of its response".
2-13	10-11	This is not a very succinct or complete description of the errors associated with modeling approaches.
2-13	12	Change to "involved *with the transformation and fluxes of nitrogen and sulfur constituents*".
2-13	14-15	It would be more helpful to be more explicit in describing the sources of uncertainty. For example with respect to deposition, wet deposition of S and N can be relatively well characterized compared to dry deposition. The major issue associated with the errors related to dry deposition are associated mostly with the modeling of deposition velocities. Also, be more specific in what is being implied with the use of the term "ecological modeling".
2-13	14-21	This section needs to include more specific information. It would be most helpful to explicitly state the major sources of error in risk estimates.
2-13	22-30	These statements are very general. At a minium specific examples need to be supplied that show actual examples of these errors and importance to the assessment.
2-14	2-3	It is anticipated that these discussions will provide sufficient detail on these sources of error in risk assessment.
2-14-15		References should also include citations to the actual literature and not rely solely on EPA documents.
3-1	10	Change to "The deposition *results*".
3-1	26	Change to "The total amount of NO_x emitted in the USA in 2002".
3-2	3-4	Change to "primary emitters of NO_x, mainly as NO and NO_2".
3-2	5	Change to "utilities, with *additional* contributions".
3-2	9	Change to "remainder of anthropogenic emissions".
3-2	10	Delete "component".

3-2	15-16	Delete "in the pie charts in".
3-2	16-17	Change to "*Results are shown on both a national* basis (contiguous states only) as well as for the eastern".
3-2	18	Delete "For this display".
3-3	1-2	Delete "Note that emissions from Alaska and Hawaii are not included in any of these charts".
3-3	5-6	(not shown) with the on-road sector *being* the largest contributor, followed by emissions".
3-3	8	Change "overall" to "*national*".
3-3	12	The figure caption needs more information (e.g. that Hawaii and Alaska are excluded in these results).
3-3	15	Change to "*contiguous* states".
3-4	3	Change to "emissions *from* fertilized".
3-4	5	Clarify what is meant by "30%–70%". Does up to 70% of the waste mass be converted to emitted NH_3?
3-4	9	Change to "impact from *the input of* total reactive".
3-4	10	Explicitly state what is meant by "This".
3-4	12	Change to "Total *USA* emissions of".
3-5	9	Change to "utilities using *coal*".
3-5	12	Change to "up to 10 *ppm*".
3-5	13	This becomes a little confusing since the figure does not provide emission data from Hawaii and Alaska. Why not include values for these states in these figures.
3-5	13-14	Sulfur is found in other forms besides amino acids in vegetation so change to "Sulfur is a macronutrient (typically being 1 to 2%) and is released as SO_2 if vegetation is combusted (Levine and Pinto, 1998)". Also, there can be other forms of gaseous releases of sulfur components from vegetation (as well as soil) including H_2S, COS, methyl mercaptan, etc. I am not sure where in the document these biogenic sulfur sources should be included.

3-6	1-2	Delete since information has been included in my suggested modification.
3-6	3-6	Change to "The *proportion* of SO_2 emissions from major sources are shown in Figure 3.1-5 both on a national total basis well as for the eastern and western United States".
3-6	6	Why not include Alaska and Hawaii for completeness?
3-6	14-16	The relative importance of this error needs to be stated explicitly.
3-7	2-5	Not sure the term "policy-relevant" is a useful term with respect to defining background concentrations. What is the basis of the actual value being used? Is it based on a specific time period or some calculation that attempts to separate out natural versus anthropogenic emissions? How can the general effects of N fertilizers and N atmospheric deposition in affecting NO_2 flux be evaluated? Is this 100 ppt based on some type of area weighted basis or some other form of calculation?
3-7	4	Delete "policy-relevant"?
3-7	25	Change to "This source distinction and quantification will".
3-8	13-14	Further information should be provided on the source and methods for estimating these deposition values. This information is provided later on page 3-13. This information should be provided earlier in the document.
3-8	19	Not sure what "Great Waters" means.
3-8	19-23	References are needed here.
3-9	Figure 3.1-6.	Reduce the significant digits in the figure legend to nearest 1/10th.
3-10	Figure 3.1-7.	Reduce the significant digits in the figure legend to nearest 1/10th.
3-11	Figure 3.1-8.	Reduce the significant digits in the figure legend to nearest 1/10th.
3-11	9-10	Further details on how these deposition estimates were derived should be provided. This information is provided later on page 313. This information should be provided earlier in the document.
3-12	Figure 3.1-9.	Reduce the significant digits in the figure legend to nearest 1/10th.

3-12-13		See previous comments on the placement of this information within the document.
3-12-16		Isn't much of this information relevant to the overall predictions of N and S deposition. It was not clear why this more extended discussion was focused on the case studies. Was there a different type of analyses done for deposition for the case study regions than for the overall U.S.?
3-16		Aren't all these abbreviations already provided and hence Table 3.2-1 can be deleted.
3-16		Are these formulas included to show the conversions from formula compound mass to nitrogen or sulfur mass? If this is there only use then Table 3.2-2 should be deleted.
3-17	11	Has the western case study been identified?
3-20		Within each of these regions there have been other measurements made of associated parameters including precipitation amount, atmospheric deposition. Is there any attempt being made in this document to compare these current results with previously published results?
3-23		There are well established geographic patterns in N and S deposition across the Adirondacks. Shouldn't these be acknowledged? There has also been substantial work on deposition estimates in the Adirondacks using other approaches. For wet only deposition for example see: Ito, M., M.J. Mitchell and C.T. Driscoll. 2002. Spatial patterns of precipitation quantity and chemistry and air temperature in the Adirondack Region of New York. Atmospheric Environment 36:1051-1062. Also other studies have evaluated the relative contribution of wet and dry deposition. See for example: Mitchell, M.J., C.T. Driscoll, J. Owen, D. Schaefer, R. Michener, and D.J. Raynal. 2001 Nitrogen biogeochemistry of three hardwood forest ecosystems in the Adirondack Mountains. Biogeochemistry 56: 93-133. Would some comparisons with other investigations be warranted?
3-25		This repeats a previous comment with respect to the inclusion of previous analyses of these regions in these cases studies.
3-26-53		There is considerable detailed treatment of the results provided by the CMAQ modeling including temporal and spatial results. With so much emphasis on these model results it would be useful to provide some other confirmation of these deposition values.

3-54	14-15	This is not entirely true. There can be instances were ambient concentrations of NO_x and SO_x are directly deleterious. These conditions, however, have not been generally found in the USA during the period of concern associated with acidic deposition effects. For Europe conditions in the "black-triangle" and other centers of high pollutant concentration there were direct impacts especially associated with SO_x.
3-55-70		The format and output of the CMAQ modeling runs are provided in some detail. Some further comparison of the CMAQ results, especially, in the areas of test cases, would be helpful in providing some objective measurements of the model results.
3-70-72		The model results need to be verified with actual data that show these relationships. Clearly the model functional relationships and parameterization will effect overall model output. It is important that it be clearly identified when model outputs are producing results that have been verified elsewhere or whether there are currently no empirical and experimental results that provide data to verify these results. Such arguments can also be made for the other N gaseous constituents.
3-70	16	It is suggested that there is "statistical imprecision in the modeling". There are other important sources of model error that need to be considered including uncertainty in model formulations.
3-82	7-9	Statements such as "Figure 3.2-55 shows that NO_x emissions account for almost all oxidized nitrogen deposition in the Adirondacks Case Study Area, while Figure 3.2-56 shows that NH_3 emissions account for almost all reduced nitrogen deposition" have important implications with respect to making recommendations associated with NO_x emission controls. Are these results verifiable and can the emissions also be linked to specific geographical areas?
3-87		How are these high emission locations ascertained? Is this a function of the location of specific monitoring locations (e.g., CASTNET sites).
3-112		This section on uncertainty (3.2.2.5) that has yet to be completed needs to include not only statistical issues, but also uncertainties in the model formulations and associated parameters.
3-113-114		This is a relatively limited reference list with considerable reliance on EPA documents. This section would be strengthened by inclusion of results from the peer reviewed scientific literature that

	support the suggested findings.
4-1 5-1	The sections on "ACIDIFICATION" and "NUTRIENT ENRICHMENT" include only outlines, but the suggestion of the importance of providing uniform terminology is encouraging.
6-1-10	This section gives a useful review of recent findings linking the cycling of S and Hg. This section also includes more references to the referred literature related to this topic
6-10-13	The section on NITROUS OXIDE provides additional information on this N gas and provides a clear indication of the importance of this gas including its linkage with N deposition and is important role as a "greenhouse" gas.
6-13-18	The synopsis on CARBON SEQUESTRATION provides a useful overview of important interactions between N deposition, warming and the carbon cycling in terrestrial ecosystems. This is a very large and important subject area. As suggested a detailed analyses is beyond the scope of this review. The term carbon sequestration does not really capture the total content of this section.
6-19-22	More emphasis in the description of aquatic effects should be placed on changes in the phytoplankton community structure that has been most noted in the western U.S.
7-1 7-15	In its current version this "Synthesis and Integration of Case Study Results" is highly descriptive. This will need to be improved with specific quantitative results that show a clear relationships to the assessment.
8-1 8-30	Clearly providing a clear scientific linkage that is policy relevant between concentrations of SO_x and NO_x and resultant S and N deposition and subsequent translation to ecosystem level effects is a major challenge. Clearly there are good linkages between concentrations of SO_x and NO_x and deposition, but the linkage to ecosystem level effects is more complicated due to inherent variability across the United States of the sensitivity of these systems to acidification, eutrophication and changes in biotic composition. It will be important to formulate an approach that takes into account these geographical patterns of sensitivity.
8-4-5	Figure 8.1-1 needs to indicate in some format that these ecological

		indicators may not be constant over the United States (e.g., N deposition levels with respect to changes biotic community structure being more sensitive in the Mountain west than the eastern U.S.). These issues are outline on lines 1-7, page 8-5.
8-5	22	The problem in not including reduced N chemical species in any formulations associated with N deposition effects is highly problematic. I am not sure it is feasible to focus solely on NO_x.
8-9		There are major difficulties in setting national standards for SO_x and NO_x concentrations without taking into account regional effects.
8-13		As suggested the development of these "functional forms" will be critical and may have both spatial as well as temporal components.
8-13		Clearly EPA is making heaving reliance on the CMAQ model suggesting the importance of validating results using other approaches and the linkages between gaseous concentrations and specific deposition levels.
8-14-17		The issues of deposition velocities are not trivial. For example look at the differences in deposition associated with the estimates at the same site for CAPMoN and CASTNET. Although CAPMoN and CASTNET provide similar concentrations of gases the deposition velocities are higher for CAPMoN verus CASTNET.
8-23		The discussion of uncertainties needs to include the various issues associated with the calculation of deposition velocities.
Attachments		
2, pg 2	5	Change "to reflect *most recent* conditions".
2, pg 2	14-15	Not sure that there is a relationship between steepness and base cation leaching rates versus the role of slope in affecting the contact of drainage waters to soils and the relative contributions of ground waters to drainage with groundwaters generally being more rich in base cations than waters derived from shallower soil sources.
2, pg 3	30	Isn't a threshold of 400 μeq/L or less considered acid sensitive? Would a threshold of 100 μeq/L better? Also, note that the correct symbol for liter is "L" not "l". Change for entire document.

2, pg. 5		Figure 3.2-1. Give a citation for the Acid Sensitive Waters (USGS).
2, pg. 7		Why was there no selection of a threshold for sulfate deposition? Is this awaiting the determination of critical load? At a minimum the European critical load could be used.
2, pg 8		Same comment for total N and S deposition and the assignment of a value as that provided by sulfate.
2, pg. 9		The inclusion of high elevation could also be justified by other criteria such as the potential contribution of occult (fog) deposition. If occult deposition accounted for in the CMAQ estimates. This also relates to rationale provided with respect topographic position (Attachment 2, page 17).
2, pg 9	25-28	This is confusing with respect S and N deposition since it was suggested above that no criteria for S and N deposition were defined. If these top quartiles are used the actual values need to be supplied that make up these quartiles. Also, the year or years from which these deposition data are derived needs to be given explicitly.
2,pg 10	14	Why "Total nitrogen (Kjeldahl)". Do you mean total dissolved nitrogen or total dissolved Kjeldahl nitrogen. Why exclude nitrate in surface waters?
2,pg 11		Although there are weak linkages between N deposition and N solute concentrations in surface waters, it would be preferable to provide the data layer of actual N solute concentrations in surface waters.
2, pg12	18-19	Provide these nutrient concentrations available from the National Nutrient Database. Provide a citation for this and the other data bases.
2, pg 15		These N deposition data layers seem to be redundant from data layers previously described.
2, pg 16		More clearly delineate the difference between "content" and concentration". Content should be reserved for the total amount (mass, molar value) of a an element on a per unit area while concentration is the amount (mass, molar value) of an element per unit of mass (sometimes) expressed as %.
2,pg 18		The delineation of the location of acidophilic lichens appears to

		have a strong boundary based upon state borders especially notable in Arizona and an absence in New Mexico.
3, pg 10		Here and elsewhere "µM" is not a correct abbreviation. The abbreviation for mole is "mol".
3, pg 12		Note that these ANC levels are lower than those suggested to be considered to be of concern in Attachment 2.
3, pg 15		There have been other models (e.g,. PnET-BGC) that have been applied particularly to the Adirondacks to evaluate especially temporal patterns of acidification. Wouldn't it be helpful to at least do some comparisons using other models besides MAGIC.
3, pg 18	8	Change to "$SO_4{}^{2-}$".
3, pg 21		Clarify how the ratios of wet to dry deposition were derived.
3, pg 22	15	Are the total deposition values used in the MAGIC calculations different from deposition estimates used elsewhere in the entire document?
3, pg 22	27-29	Some elaboration on how ASTRAP derived deposition would be helpful and how these estimates confirm or differ with the CMAQ estimates.
3, pg 23-26		There is considerable discussion of the calibration of MAGIC in this section. Some of this could be reduced and inclusion of other ways of comparing model output with other published results including other models (e.g. PnET-BGC) would provide additional perspectives of these results.
3, pg 29		Similar to previous comments, although the discussion and definition of the F-factor provides background information this is not a novel approach, but rather one that could be cited. More emphasis of the validation and comparisons with actual measurements versus description of the model development would be more helpful.
3, pg 31		Figure 5.1-1. Trends in LTM monitored lakes in the Adirondacks of New York would be improved if specific lake classes were used. It is not clear from these figures how a general trend for the LTM monitored lakes were obtained.
3, pg 32		Figure 5.1-2. The modeled values appear to be substantially different that those of the measured values. Doesn't this bring into

	question to validity of the predictions for 2010? I don't believe the results of this figure are discussed within the document.
3, pg 35	Clearly the document is incomplete at this stage.
4, pg 1-2	This seems repetitious of information provided in the main document.
4, pg 5	"M" is not the correct SI abbreviation for mole. The correct abbreviation is "mol".
4, pg 4-9	Not sure that this much restating of what has previously been found about Al and Ca relationships is needed.
4, pg 11	Would it be more efficient to cross reference the description of ecosystem services as provided in the main document?
4, pg 18-20	Are the details of the history of the Kane Forest needed in this document?
4, pg 21-25	Similarly to the previous comment, are all these details about Hubbard Brook needed for this document?
4, pg 26-47	This is difficult to follow due to the level of detail. Perhaps a more generalized format in which could be imbedded specific references to the literature or the inclusion of appendices so that the major theme of the discussion is not lost.
4, pg 50	Not sure of the importance of the inclusion of the section on "Implications for other systems" in this document.
4, pg 51-52	The uncertainties issues need to be better integrated into the overall document.
4, pg 53	The "Conclusions" summarize the important issues and these issues should be the focal points of this entire section with a need
6, p 43	The figure on this page (Figures 5.1-1 5.1-2) appears to have an inappropriate numbers. Shouldn't they be 6.1-1 and 6.1-2?

Mr. Richard Poirot

These comments pertain primarily to REA Chapter 3 (Sources, Ambient Concentrations, and Deposition), for which the following charge questions were provided.

> **Question 1**: To What Extent are the air quality characterizations and analyses presented in chapter 3 technically sound, clearly communicated, appropriately characterized, and relevant to the secondary NAAQS for NOx and SOx?
>
> **Question 2**: Section 3.2.1 describes an approach for evaluating the spatial and temporal patterns for nitrogen and sulfur deposition and associated ambient concentrations in the case study locations. This draft document includes the analysis for the Adirondacks Case Study. <u>*Does the Panel agree with this approach and should it be applied to the other Case- Study Areas?*</u>
>
> **Question 3**: Section 3.2.2 describes the relative contributions of ambient emissions of nitrogen and ammonia to nitrogen deposition for the case-study areas. <u>*To what extent is the approach taken technically sound, clearly communicated, and appropriately characterized?*</u>

Assuming the many and important missing "placeholder" sections are filled in, I think the air quality characterizations and analyses presented in chapter 3 will provide a technically sound, clearly communicated, appropriately characterized, and relevant to the secondary NAAQS for NOx and SOx (although it still remains unclear to me what kind of 2ndary standards are being contemplated). Many of these placeholders refer to evaluations of model performance, and since the majority of the chapter consists of presentation of intensively graphical model results, filling the placeholders will be key to providing confidence in the model results. The technical approach appears to be reasonable, but without a better sense of model performance (including both CMAQ and the RSM meta-model), the technical soundness and relevance to NAAQS can't really be evaluated.

The presentation of the information is clear (even beautiful), but there is minimal discussion or interpretation of the graphical results, which makes it difficult to maintain interest in looking so many pictures. Conversely, it might be informative to poll the model results in ways that might provide a better understanding of relationships among the various metrics of air quality, deposition and environmental effects. For example: what would the maps look like that show the ratios of S deposition to S emissions; N (oxidized) deposition to N (oxidized) emissions; N (reduced) deposition to N (reduced) emissions. What would maps look like that show ratios of S (or N) deposition to ambient SO_2 (or NO_2) concentrations? What if modeled S emissions were rolled back to show alternate lower maximum levels of SO_2 (and/or NO_2) – what would be the subsequent changes in S (and/or N) deposition in sensitive downwind areas? If sections of the Adirondacks were considered to be experiencing adverse levels of acidifying deposition, how could the models be used to determine the "significant contributing area" – or perhaps some combination of emission size, and frequency upwind – such that a non-attainment area might be defined to better include the contributing emissions? A possible approach for an improved secondary SOx or NOx NAAQS might consider (much) lower levels but averaged over larger areas or longer averaging times.

With the caveat that the model performance needs to be more clearly examined and documented, I think the more detailed approach applied here for the Adirondack case study is reasonable, and could be applied to other case study areas. One possible concern – possibly more relevant to the Adirondacks than to most of the other case study areas – is whether the 12 km gridding might be coarse relative to the spatial variability in deposition, terrestrial & aquatic ecosystems and associated effects. Potentially the larger grid cells may fail to adequately capture orographic increases in precip volume & deposition and the additional increases from occult deposition at highest elevations (or in certain coastal areas) – in comparison with variations in sensitive terrestrial and aquatic biota within these grid cells . See for example: Miller, E.K. et al. 1993. Atmospheric deposition to forests along an elevational gradient at Whiteface Mountain, NY USA. Atmos. Environ. 27A:2121-2136.

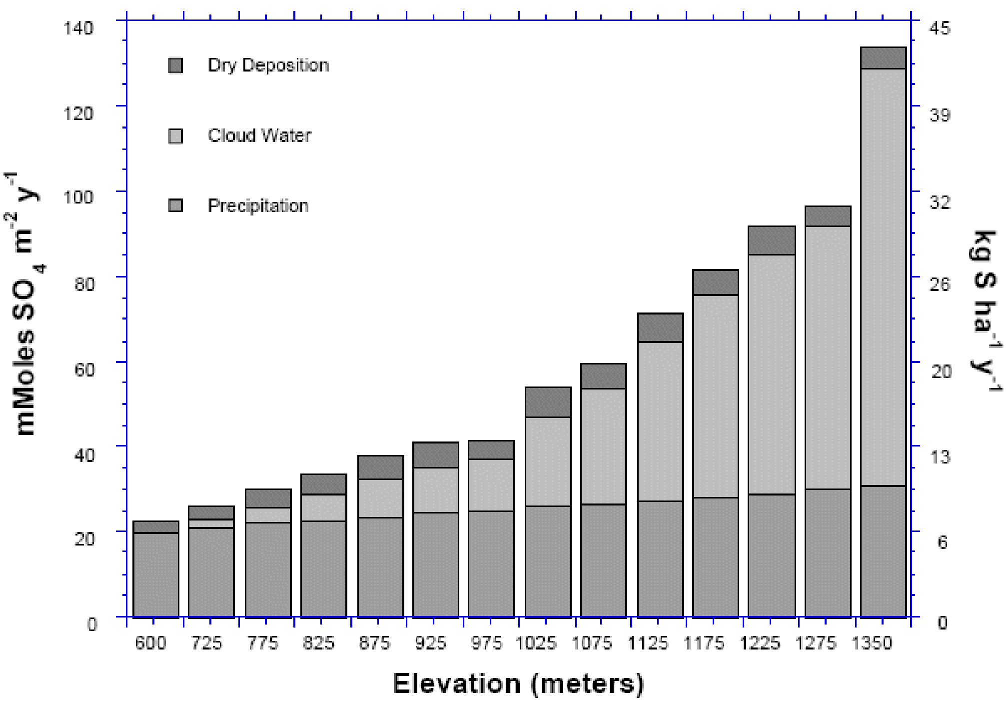


Figure 1. Variation in wet, dry, cloud, and total sulfur deposition over ~2km ground distance as a function of elevation on Whiteface Mt., NY, 1986-1989 (from Miller et al. 1993).

Possibly this influence of occasionally large terrain, deposition and species variations within the relatively large grid cells could be evaluated by conducting a higher resolution sensitivity analyses within selected grid cells in the Adirondacks case study area. Alternatively, it might at least be possible to disaggregate the coarser modeled dry dep. estimates to combine with the higher resolution wet dep. estimates from the Grimm & Lynch approach. Arguably the dry dep totals and variability may be less important than wet dep at more remote receptor locations. Possibly also a terrain-based cloud deposition model could be added to provide added detail for higher elevations or in coastal areas.

Regarding the soundness of the technical modeling approach to estimate the relative contributions of ambient emissions of oxidized and reduced nitrogen to nitrogen deposition for the case-study areas – the approach seems sound but again there is a need to evaluate the performance of the models (CMAQ + RSM). Trust but verify. This evaluation could/should extend where possible to subcomponents of the models such as emissions inputs and space/time patterns of modeled estimates of wet oxidized N and wet reduced N deposition.

That being said, I do like the approach of directly linking the estimates of changes in Nr deposition to changes in specific emission sources. I would think a similar approach could also be taken to zero out and otherwise reduce SOx emissions from different source categories. Down the road, this may allow a bundling & comparing of projected "optional" future emission controls (small, medium, large, etc.) to desired reductions in deposition in sensitive areas, in comparison to the associated concentrations in atmospheric NOx & SOx concentrations (in the event the layers say we have to stick with the traditional indicators). What new NAAQS limits of these indicators (perhaps averaged over larger - or much larger - areas than single monitors) would be necessary to achieve the desired reductions of Nr and/or S deposition in sensitive areas?

In addition, if you were to add various levels of SOx (and NOx) emission reductions to your modeled scenarios, it would be a snap to calculate and display the resulting changes in sulfate and nitrate aerosols (and in their visibility effects) that would result from any changes in S & N emissions or deposition or SO_2 & NO_2 concentrations. This would allow you to (a) consider a more complete set of welfare benefits that would result from any revised NAAQS based on deposition effects and/or (b) might lead to and help justify alternative ambient air indicators – for example the sum of total atmospheric oxidized S and N compounds (sum in ug/m3 of S from SO2 and pSO4 and N from NO, NO2, HNO3, and pNO3 or somesuch) – that might not be considered if only deposition-related effects are considered, but which might, set at the right levels, result in large deposition-related benefits.

Other Minor Comments:

p. 3-1 lines 16-23: This is a good example of what seems like an intentional sense of vagueness regarding which pollutants and secondary transformation products are or are not the subject of this review, and/or available as potential indicators for secondary NAAQS. Why are nitric acid and PNO3 considered part of "NOx", while SOx includes only gaseous SO_2?

p. 3-4, Figure 3.1-3: This figure just doesn't look right – and seems inconsistent with the reduced nitrogen deposition map in Figure 3.1-8 on p. 3-11.

p. 3-4, lines 3-5: Does confined feeding really increase animal wastes, or does it just increase atmospheric emissions from them?

p. 3-4, line 12: Figure 3.1-4 does not show annual 2002 SO_2 emissions "by state".

p. 3-5, line 4: I would change "Industrial" to "Anthropogenic".

p. 3-5, lines 10-15: Should marine DMS emissions be mentioned here?

p. 3-6, lines 14-16: Can you provide any quantitative indication of how large these fire SO_2 emission underestimates are?

p. 3-8, lines 5-7: Does this increased deposition of reduced nitrogen also pertain over the 100-year period of the preceding sentence. Can you say something more precise about trends over the past several decades?

p. 3-12, lines 10-14: This description relates to how NADP interpolates its wet deposition data. However, later you indicate using the precip-volume-enhanced estimates from Grimm & Lynch (2004). So which was it? One general concern is that the 12 km gridding may overly smooth some of the more extreme orographic increases in precip – and you seem to exclude cloud water deposition. Possibly this could be handled by conducting some sensitivity analyses for grid cells containing higher elevation terrain – especially for the Adirondack and Shenandoah case studies.

p. 3-16, line 11: Why not also describe CASTNet (& AIRMoN dry) data for HNO_3, NO_3, NH_4, SO_4?

p. 3-23, lines 4&5 and lines 11&12: say roughly the same thing twice.

p. 3-23, line 7: "(25% wet vs. 6% dry)" and 69% what?

p. 3-23, line 12: Change "does" to "do".

p. 3-54, line 17: You could add "current and historical" before "atmospheric deposition".

p. 3-55, lines 20-21: Can you provide any indication of if and how well the RSM technique (and for that matter the underlying CMAQ model) works for all the SOx, NOx and reduced N species you will use it for?

p. 3-57, line 5: Am I missing something or did you only use a couple (zero-outs) of these 210 control runs? So what was the purpose of the other runs?

p. 3-71, line 8: Delete one "deposition" in "greater deposition of oxidized nitrogen deposition".

p. 3-71, lines 8-15: I would think formation of aerosol NH_4NO_3 would tend to increase the transport distance, but would not lead to any net decrease or increase in Nr deposition. It all gets deposited eventually. An exception might be if aerosols persist long enough to transport out of the (US or North American) domain.

p. 3-73: I must have blinked somewhere, because I didn't expect to see this large, international "sugar maple case study" area discussed previously. Its not listed as a "Case Study Location" in

Table 2.1-1 on page 2-2&3. It reminds me to ask for a clearer “up-front” description (& map) of all the intended case study areas.

Mr. David Shaw

General Comments

Thank you for addressing the outcome of this REA. I believe that this makes the document more focused.

I still feel that a specific goal of this assessment should be to identify and report sensitive areas which do not have adequate monitoring. Adopting this longer view in this analysis will enable the next review process to start from a stronger point. In identifying areas without adequate monitoring data, we may be able to start the process of getting a stronger data record of results for future analysis. It is my hope that this will lead to better modeling due to better data.

While there are certainly areas that are deficient in monitoring data, there definitely are areas of strong monitoring data with analysis. I still feel that the areas of certainty do not receive equal treatment as uncertainty. I feel we must be clear that we do understand causes, effects and variability in our ecosystem response.

On this note, the analyses presented in Chapter 3 of the REA rely heavily on modeling and don't always reinforce where the measurements are the strongest. It is important to emphasize where we have the most confidence (e.g. wet deposition, ANC measurements in case study areas) and the least confidence (e.g. air concentrations and dry deposition of NOx/SOx, where measurements are lacking). Much of the information on dry deposition will come from CMAQ, and the measured data from CASTNet and other special studies could be used to assess the model at selected locations.

Charge Questions

Scope of the Review
Question 1

Chapters 1 and 2 provide the background, history, and the framework for this review, including a discussion of our focus on the four key ecological effect areas (aquatic acidification, terrestrial acidification, aquatic nutrient enrichment, terrestrial nutrient enrichment). Is this review appropriately focused in terms of the targeted areas and in terms of characterizing the important atmospheric and ecologic variables that influence the deposition and, ultimately, the ecologic impacts of nitrogen and sulfur? Does the Panel have any further suggested refinements at this time?

Pages 1- 1 to 1-10:
I feel that this gives good overall information on the Rational, Background and History. It might also be a good place to address other pollutants associated with NO_x and SO_x analyzing the whole set of problems associated with these pollutants.

Page 1-8 lines 3-5:
Lines 3-5 state that 'in spite of the complexities and uncertainties….' it became clear that a program to address acid rain was needed. In actuality, it was the evidence of a preponderance of the scientific data of the NAPAP effort which made it clear that an acid rain program was needed. This builds on one of my general comments regarding an emphasis on uncertainty rather than giving equal treatment to those areas where certainty exists.

Page 1-16:
I would recommend adding a discussion on critical loads as the organizing principle of this RAE assessment. Include the current understanding of ecological indicators and how levels of the proposed standard will be integrated.

Page 1-20:
It would be helpful to explain how the existing monitoring data will be used to evaluate the success of the proposed standard? How and when will the existing monitoring networks be evaluated for adequacy of measuring ecosystem response?

Page 2-10:
I appreciate the effort in adding sulfur and mercury methylation. The fact that it will be addressed will make this a stronger analysis.

Page 2-11, Table 2.3-1:
I feel that it would be beneficial to add to Cultural Services "chemical and biological degradation of Constitutionally (federal and states) protected Wilderness areas". This would apply to several if not all Targeted Effect Areas. The point being that the ADK case study area, for example, represents a 6 million acre region, 43% of which is protected by the NYS Constitution as 'forever wild' Forest Preserve. Here, tree cutting is not allowed, yet atmospheric deposition damages forests and diminishes aquatic ecosystems within these forests. The remaining 57% is devoted principally to forestry, agriculture, and open-space recreation, a portion of which is sensitive to negative effects of atmospheric deposition. This landscape holds an additional cultural value to New Yorkers especially, and to others from the US and around the world.

Pg 2-12:
In the box where "provisioning services" and "cultural services" for Sulfur and Mercury Methylation are provided, fish kills is listed as an ecological impact. From the literature that I am familiar with, fish kills or declines in fish populations are not a good indicator of MeHg, however, declines in the success of species higher in the food web such as loons and humans occur because of MeHg neuron toxic effects and bio accumulation.

Air Quality Analysis
Question 1

To what extent are the air quality characterizations and analyses presented in Chapter 3 technically sound, clearly communicated, appropriately characterized, and relevant to the review of the secondary NAAQS for NO_x and SO_x?

Sections 3.1.4 and 3.2:
It seems as if the dataset discussion (Section 3.2) would be more beneficial if it appeared before the composite deposition maps (Section 3.1.4). These composite maps consist of modeled dry and measured/interpolated wet deposition. For these maps, are the units actually kg ha^{-1} y^{-1}, or are they kg N ha^{-1} y^{-1} and kg S ha^{-1} y^{-1}? Some mention should be made that the dry deposition estimates are generally consistent with whatever measurements (e.g. CASTNet) are available, that one year of modeling is adequate to capture the seasonal/spatial variability in the predictions (i.e. this is not an atypical year), and that other photochemical models are capable of estimating dry deposition and could be used to perform such an analysis.

Section 3.2.1.2:
I am interested to know why SO_2 and NO_2 from SLAMS/NAMS monitors are not included in the measured database? While there may not be many NO_2 monitors in rural areas, there should still be some measurements of SO_2 in the case study areas.

Page 3-21 ADK case study area:
There are several questions regarding the selection of this study that would be of interest to note in the REA:
How were the 44 lakes and ponds selected?
How was the subset of 15 lake sites selected for geographic variation in deposition assessment?
How representative are these sites of the whole region?
Is there an elevational stratification?
Are any of the intensive study sites part of the current Adirondack monitoring programs (e.g., ALTM, TIME, AEAP)?
Have any or will any of the model results be compared with existing long term monitoring data?

Some important facts to point out regarding this region is that 22% of it is above 600 m were the sensitive spruce fir forest community becomes dominant. Further up above 900 m are key signature mountain peak ecosystems containing over 100,000 acres.
Monitors are not measuring any deposition data above 610 m in NYS, with the exception of the top of Whiteface Mountain.

Pages 3-35 through 3-37, and 3-50 through 3-53:
It may not be necessary to include the additional information on a monthly basis. The form of the annual NAAQS looks to be seasonal or annual, so presenting deposition on a seasonal basis seems to be adequate to capture the variation over the course of a year.

Figures 3.2-29 through 3.2-44:
I would recommend that the metrics for these figures are clearly stated. Also, it would be helpful to confirm that the "whiskers" are the minimum and maximum and that the boxes are $25^{th}/75^{th}$ percentiles.

Figures 3.2-45 onward:
It might help the reader to reverse the color scheme, that is display the smallest impacts in green and the largest impacts in red.

Misc.

In the introduction there are many references to "…noted ***below*** in Section 1.x…." I would recommend removing the word "below" in each of these references.

Page 1-3 line 19, "Chapter 2" should be

Dr. Kathleen C.Weathers

Overarching comments on Chapters 7 and 8:

First, I think that the case study approach is useful and appropriate for this analysis. Next, Chapter 8 provides some interesting ideas to ponder. Finally, the incompleteness of this document makes it difficult to provide much in the way of pithy comments. In addition, there's much editing that will need to be done on these chapters.

Case Studies: As we discussed during the first meeting of the Panel, the choice of case studies is critical. My biggest concern about the case studies identified in this draft is whether they are representative enough. That the there is sufficient data and information for a case study is, of course, a major criterion. Further, their use should be supported and justified in regard to the suite of impacts of air pollution they illustrate with regard to direct and indirect effects of atmospheric N and S. I question the use of the Chesapeake Bay/Pamlico Sound in this regard, mainly because atmospheric N deposition to the region is not a large proportion of N inputs to the ecosystem. Thus, unless it is used as a contrasting example of a system in which the atmospheric deposition of N is not a major component of N loading (I note that this contrast could have utility), the Chesapeake Bay/Pamlico Sound regions may not be the most illustrative case study to use. I do think that, if feasible, evaluating deposition impacts on a coastal ecosystem(s) would be instructive.

I mentioned in my last review that I thought Day-Cent-Chem should be considered as a potential model to be used for these case study analyses. It has some advantages over Magic, and some parallels with other biogeochemical models (PNET-BGC, for example).

Would it be possible to consider in some creative way a "case study" that summarizes and synthesizes experimental manipulations of N and S inputs (e.g., Harvard Forest, Catskill Mountains, NY, Bear Brook, ME, Mt Ascutney)?

Conceptual pieces: I agree with using a "weight-of-evidence" approach, which I interpret to mean that assessing effects using a suite of different tools (modeling, experiments, theory, long-term monitoring) can be used to provide the most robust syntheses. The IPCC has used this approach successfully.

As the authors of this document are no doubt well aware, ecosystems are affected by the amount (and sometimes concentration) of a pollutant or nutrient that is actually makes its way to the ecosystem — i.e., that is deposited. Working backwards from that fact suggests that what will be necessary is a set of standards that consider actual loading — deposition — to ecosystems, rather than atmospheric concentration-based standards that identify the role of "modifying factors." The devil's in the details of Figure 8.1-1! As research has demonstrated, for some chemical species, there may be little connection between emissions and total deposition downwind, much less all of the biologic and geochemical transformations that happen from deposition to terrestrial processing to surface water effects (or not).

As noted in this REA, lining up the spatial and temporal scales of analysis and output are critical issues, for example, high temporal resolution (air concentrations and deposition on an hourly basis, for example) but low spatial resolution (across the country) may be exactly the opposite of what would be most useful for assessing ecosystem effects.

The likelihood that current rates of deposition will show immediate effects on downstream ecosystems for nutrients such as N is quite low. Rather, capturing the longer-term average loads and patterns of deposition is likely to be more relevant.

The derivation of deposition metrics and transformation functions were at once described in great detail and in not enough detail. Seeing the forest for the trees will be very important. See comments about a deposition standard, above.

General comments on Chapters 7 and 8:
Note that I have not offered detailed editorial comments for this review.

I'm not sure that I agree (page 7-2, lines 15-16) with the statement that it is the increase in sulfur and nitrogen content of soils that is the sole reason for alteration of major biogeochemical processes. Also, just below, missing from the description of acidic deposition's effects on aquatic system are the properties of the adjacent terrestrial watershed, including hydrologic pathways. And, at the end of this paragraph, the reasons why the results from the case studies for the Adirondacks and Blue Ridge Mountains might be applicable to the rest of the Appalachians and the Northeast should be identified.

There appears to be some confusion about terrestrial acidification, or at least the beginning of the terrestrial acidification should be rewritten for completeness (i.e., natural and anthropogenic acidification occur in soils), accuracy, and clarity.

I appreciate the importance of the statements on pages 8-12, lines 9-13 in regard to a national transformation function with local modifying factors within it. This is very important given the importance of landscape (in its broadest sense) in influencing inputs.

I note the absence of cloud or fog deposition in Section 8.3.2.1. I suggest mentioning in the text that it will not be included directly here. This vector can be critically important, but only in coastal and mountainous ecosystems.

Surely the intention in the Catchment parameter section is to use GIS databases (not actual topographic maps)?

Be sure to label figures clearly as well as be clear when discussing total (wet + dry? or Nr, or?) deposition. Note annual or seasonal as well in the text.

Organic nitrogen, in regard to both its atmospheric input (less known) and output (more known) via surface waters should be at least mentioned as part of the N effects picture. While it is not directly part of NOx/SOx emissions, it is nonetheless important and it is becoming especially relevant in regard to climate change.

Such terms as ecosystem services, and ecosystem health, should be explicitly, consistently, and clearly defined among and within these NAAQS review documents.