UNITED STATES ENVIRONMENTAL PROTECTION AGENCY
WASHINGTON D.C. 20460

OFFICE OF THE ADMINISTRATOR
SCIENCE ADVISORY BOARD

August 28, 2009

EPA-CASAC-09-013

The Honorable Lisa P. Jackson
Administrator
U.S. Environmental Protection Agency
1200 Pennsylvania Avenue, N.W.
Washington, D.C. 20460

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

Dear Administrator Jackson:

The Clean Air Scientific Advisory Committee (CASAC) NO_x & SO_x Secondary NAAQS Review Panel met on July 22-23, 2009 to review EPA's *Risk and Exposure Assessment (REA) to Support the Review of the Secondary National Ambient Air Quality Standard for Oxides of Nitrogen and Sulfur: Second Draft*. This letter has been reviewed and approved by the chartered CASAC at a public conference call on August 20, 2009. In this letter, CASAC offers general comments and recommendations for finalizing the REA. We also address the Agency's framework for developing the forthcoming Policy Assessment. During the meeting, EPA's Office of Air Quality Planning and Standards (OAQPS) informed the Panel of the final schedule for the current primary and secondary NO_x and SO_x NAAQS reviews and rulemaking. CASAC is very concerned about the schedule's implications on the policy options under consideration by the Agency for the secondary standards.

EPA staff informed the Panel that there is insufficient time to meet its court-ordered deadlines and complete the level of analysis needed to sufficiently formulate and justify recommendations for a secondary NAAQS for NO_x and SO_x. EPA will likely propose one of three alternatives for rule-making:

1. Retain the current secondary standards;
2. Revise the current NO_2 secondary standard to make it identical to the primary and retain the current secondary standard for SO_2; or
3. Revoke the current secondary standards for NO_2 and SO_2.

None of these options represent a NAAQS(s) with the appropriate indicators, ambient levels, forms, and averaging times to address secondary, welfare effects. The Integrated Science Assessment (ISA) and the REA both provide ample evidence that deposition of oxidized forms of sulfur (SO_x) and nitrogen (NO_x), along with chemically reduced (NH_x) and volatile organic forms (RH_x) of reactive nitrogen (Nr) adversely impact sensitive ecosystems at current ambient atmospheric concentrations. All three of the proposed options leave at risk sensitive aquatic and terrestrial ecosystems in many parts of the United States.

Previous CASAC panels and National Research Council Reports have called upon EPA to do exactly what is being attempted in the current NO_x and SO_x reviews: develop standards that appropriately integrate across pollutants and outcomes. The Panel recognizes the complexities involved in developing the information needed to support an ecologically relevant, multi-pollutant standard(s). Given that CASAC has no purview to grant the Agency more time, we recommend the Agency complete the current reviews for NO_x and SO_x to fulfill its statutory and legal obligations, and simultaneously expedite the analyses to support ecologically appropriate welfare standards. The Panel supports a fast-track development of an alternative Policy Assessment (and the associated rulemaking). Building upon the ISA and REA completed in the current review, EPA should complete the expedited analyses in less than two years.

With regard to the review of the second draft REA, the Panel found it greatly improved over the prior version and responsive to CASAC's advice. EPA staff is commended for providing the information essential in development of a set of secondary standards to address multiple ecosystem outcomes resulting from the exposure to multiple pollutants, some of which are not criteria pollutants. The Panel considers the following issues critical to finalizing the REA:

- The Executive Summary (ES) is still not a comprehensive summary of the REA. The Panel maintains that the ES is a very important part of the REA. In its current form, however, it does not fully capture the conclusions and findings presented in the REA.
- The synthesis of case studies in Chapter 7 is inadequate. The chapter provides a summary of the case studies, but does not fully synthesize their results. Nor does Chapter 7 provide the type of contextual information that is needed to extend these results to broader national scales.
- The treatment of uncertainty in the current draft REA should be improved to strengthen the analysis in this review. The application of the various steady state, dynamic, and statistical air quality and ecosystem process models is the foundation of the REA. The Panel recommends the addition of a model performance evaluation to describe the strengths and limitations of the various models used, especially with respect to spatial and temporal predictions, and how the models can be effectively used to inform the decision-makers about the time needed for ecosystem recovery. A table summarizing model characteristics and capabilities would be especially useful in the ES.
- The Panel would also like the REA to acknowledge the potential benefits of reactive nitrogen deposition on nitrogen deficient ecosystems.

The CASAC and Panel membership is listed in Enclosure A. The Panel's consensus responses to the Agency's charge questions are presented in Enclosure B. Individual review comments from the Panel are compiled in Enclosure C. We look forward to the Agency's response and the opportunity to provide advice on EPA's Policy Assessment document.

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

Rosters of the NO_x & SO_x Secondary Review Panel and CASAC

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CASAC NO_x & SO_x Secondary Review Panel

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NOTICE

Enclosure B

CASAC Panel's Consensus Responses to EPA's Charge Questions

Executive Summary:

1. *In response to the Panel's review of the first draft Risk and Exposure Assessment, we have included an executive summary of this document. Does the Executive Summary adequately summarize and characterize the key issues driving this review as well as the important findings of the analyses? Does the Panel have any suggestions for clarification or refinement of the Executive Summary?*

The Panel is pleased to see an Executive Summary (ES) in this draft of the REA, which was recommended in the Panel's review of the first draft. However, the ES is still not a comprehensive summary of the REA because it fails to capture the major findings and conclusions presented in Chapters 3 – 7. One way to highlight the important results of the REA would be to replace the current "Conclusions" section with one entitled "Key Findings", similar to the format of the ES in the ISA. In both the ES and Chapter 1 a number of important policy-relevant questions are listed. It should be explained in the ES that these questions are not answered in the REA; rather they set the stage for their discussion in the forthcoming policy assessment document. Also, the list of policy-relevant questions in the ES is not consistent with the list in Chapter 1. It would be helpful if the sequence of questions followed the logical layout of Fig. ES-2.

The introduction to the ES should explain why the review focuses on the ecological effects of NO_x/SO_x deposition, but takes a limited view of other potential welfare effects such as foliar injury from gaseous phases of NO_x/SO_x and other effects of deposition such as injury to materials. An explanation of the selection of the case studies is also missing from the ES. The reader is left to ponder if the justification is based on data availability, representativeness, sensitivity, or some combination of these criteria. The ES should provide a sense of what percentage of the US exhibit similar problems as represented in each of the four categories of the ecological effects. The treatment of reduced forms of nitrogen (NH_x) is much improved in this second draft. However, the regional nature of ammonia and NH_x needs to be clearly acknowledged in the ES and throughout the REA. One way to illustrate the regional nature of nitrogen deposition would be to supplement Fig. ES-7 with two new figures, one for NH_x and the other for oxidized N.

The key data gaps should be identified in the ES, along with EPA's intention/recommendations to fill those data gaps. The Panel was pleased with the inclusion of an explanation of the concept of ecosystem services and how this concept may ultimately help to quantify adverse effects on public welfare. However, if specific examples such as recreation usage are included in the ES, these must be tied, at least qualitatively, to the effects of deposition on the quality and abundance of these ecosystem services. The Panel maintains that the ES is a very important part of the REA and, in its current form, does not fully capture the conclusions and findings presented in the REA.

Air Quality Analyses (Chapter 3):

1. *This chapter describes an approach for characterizing the spatial and temporal patterns of nitrogen and sulfur deposition in the case study locations including both oxidized and reduced nitrogen, and both wet and dry deposition of oxidized nitrogen, reduced nitrogen, and sulfur. Are the uncertainties associated with these analyses appropriately identified and described?*

The Panel was encouraged to see that the Chapter 3 characterization of emissions, air quality and deposition information was a substantial improvement over the previous draft, especially in combination with the Appendix 1 performance evaluation of the CMAQ model that is prominently employed in the air quality and deposition estimates. The detailed map presentations of the various emissions, air quality and deposition patterns are informative and directly responsive to prior CASAC requests.

A relatively complete "listing" of uncertainties associated with the air quality-related characteristics is provided in section 3.5. However, the discussion is minimal and the absence of any quantitative estimates of uncertainty substantially limits the usefulness of this section. The Panel recommends that some quantitative estimates of uncertainty be provided here, even if these are not intended to be comprehensive. For example, the (informative, but graphic-only) comparison of various model and measurement-based sulfur deposition estimates for the Adirondack case study area could be enhanced by scatter plots which show quantitative differences among the estimates. Some of the CMAQ model performance metrics from Appendix 1 might also be summarized in Chapter 3, along with a comparison of results from CMAQv4.6 vs. CMAQv4.7. At a minimum, some indication should be provided of the relative uncertainties of the different air quality and deposition estimates, which are presumably greater for dry than for wet deposition and greater for NH_x than for SO_x. For example, EPA has done some work on quantifying uncertainties in ammonia emissions that could be included here.

2. *In response to CASAC's recommendation, the RSM analysis presented in the first draft Risk and Exposure Assessment was replaced by an analysis of results from a new series of CMAQ simulations designed to explore the relative contributions of NO_x and NH_3 emissions to total, reduced and oxidized nitrogen deposition and the relative contribution of SO_2 emissions to sulfur deposition. Does this approach enable us to adequately examine the contribution of NO_x to total nitrogen deposition?*

It would also be informative to see a number of "ratio" or "difference" maps. For example, maps showing the ratios of wet, dry and total sulfur deposition to SO_2 concentration and to SO_2 emissions, and ratios of wet, dry and total N, and oxidized N deposition to NO_2 concentrations and to NO_x emissions would be informative. Ratio or difference maps would also be an informative way to present additional details on CMAQ model performance. For example, what is the ratio of or difference between CMAQ modeled wet S (or N) deposition and measured (interpolated NADP) wet S (or N) deposition?

Some explanation should also be given for why potentially important contributions to NO_x emissions from lightning and soils and from increased S and N deposition from cloud water at higher elevations were excluded from the analyses. The Panel feels there is merit to include some discussion of the potential importance of the effects from these sources.

3. *The CMAQ application and model performance evaluation is presented in Appendix 3-1, as recommended by the Panel. Is this analysis sufficient to support the use of the model in this review?*

The evaluation of CMAQ model performance in Appendix 1 is a substantial improvement over the previous draft, and provides added confidence in some of the deposition estimates, their inter-annual variability and their uncertainties. However, the Panel recommends that additional model performance metrics be shown to fully assess the model performance to gain confidence in applicability of CMAQ for this assessment. These metrics should include model performance statistics (and plots) for daily or weekly averaged quantities (based on available data), mean normalized bias (and error) instead of normalized mean bias, and model performance for specific regions rather than the whole country. It would also be useful if model performance for NO_x is also included whatever such data are available. There should also be more discussion of what level of model performance is considered "acceptable", as there were many results for which the model performance were not particularly compelling.

Case Study Analyses (Chapters 4 & 5):

Questions related to the individual case study analyses are presented below. Overarching questions across all the case studies include:

1. *Are uncertainties appropriately characterized across the case studies? Is there adequate information to allow us to weigh the relative strengths of each case study to inform the standard setting process?*

The discussion of uncertainty was generally well presented in a qualitative sense, but needs to be made more quantitative wherever possible. In particular, it would be useful to convey a sense of the relative importance of the various sources of uncertainty and articulate which are critical for policy assessment. This should include a discussion across the various links in the proposed structure of the standard (Fig. ES-2 and throughout); i.e., consider the impacts of uncertainty in CMAQ inputs and outputs on deposition, and the subsequent impacts of deposition estimates on ANC, and then the impact on critical loads. It is difficult to assess the importance of this uncertainty due to inherent problems in extrapolation from the case studies to other ecosystems or across larger regions. Some of this uncertainty is related to the case studies because they do not represent the full spectrum of effects associated with the deposition of sulfur and total reactive nitrogen.

2. *In using the Risk and Exposure Assessment to inform the policy assessment, we plan to focus on aquatic acidification as the basis for an alternative multi-pollutant secondary standard as this is the area where we have the most confidence in our ability to characterize adverse effects. Does the Panel agree with this approach?*

The Panel agrees with the focus on aquatic acidification, based on the quantity and quality of data available, but strongly recommends that EPA continue to consider multiple indicators and find a way to truly integrate multiple indicators into a standard. Multiple indicators will expand the geographic relevance and scope of the standard and afford protection to a maximum number of sensitive ecosystems. Moreover, recent findings suggest that the recovery of soils from acidification may take an extended period, which over the long-term will also affect the rate of recovery of aquatic ecosystems.

Chapters 4 and 5 should also include an overview of the tools and models used in the case studies and present a justification of EPA's choices, in particular with respect to the use of SPARROW and the choice of BC/Al ratios rather than the more commonly used Ca/Al ratios. An additional case study or at least a discussion of a nitrogen-deficient site is needed in order to avoid potential bias in extrapolating from only sensitive sites. Additional discussion on aquatic-terrestrial linkages would be helpful, as well as explicit information on the relationship between deposition and ANC.

Acidification:

The CASAC is generally pleased with the effort to evaluate the effects of acidification on aquatic and terrestrial ecosystems. However, this chapter needs considerable editorial work including the use of proper terminology, reduction of duplication and improvement in the clarity of the text and figures. A more accurate use of concepts is needed and terminology should include descriptions related to ANC and buffering. The "critical loads" concept is important, but the text needs to clarify its utilization both in published literature and its adaptation in the REA. Averages and steady-state calculations are used in various ways including the application of critical loads and determining the effects of ANC. The advantages and limitations of using averages and steady state calculations need to be provided. In the REA, the effects of sulfur and nitrogen deposition needs further description to delineate the relative importance of natural acidification and internal ecosystem processes in affecting the temporal and spatial patterns of acidification.

1. Section 4.2 and Appendix 4 describe the analyses used to evaluate the effect of aquatic acidification. The analysis evaluates the ANC in selected lakes and streams in the Adirondacks and Shenandoah relative to three potential ANC cutoff levels (20, 50, and 100 µeq/L) to determine the impact of current levels of deposition in these areas as well as a larger assessment area. Are these data adequate to establish critical loads of deposition for the case study area?

The use of ANC to evaluate the impacts of current levels of deposition is appropriate. The emphasis on using the results from aquatic ecosystems for analysis and model predictions on acidification is appropriate based upon available historical and temporal data sets. The implementation and uses of models (e.g., CMAQ, MAGIC, SSWC) are integral to this chapter and the focus of the REA in ascertaining the effects of acidification. The analysis of uncertainty within models focuses mostly on variation in parameter estimates and how this variation affects model output. Further discussion on why specific models were selected and the implicit limitations (e.g., processes not covered, representation of internal elemental cycling, appropriateness of spatial and temporal scales, etc.), as well as the strengths of the various models would help the reader evaluate the appropriateness of model results and applicability for predicting acidification temporal and spatial patterns of acidification.

2. The ecological effect function for aquatic acidification (section 4.2.7) attempts to characterize the relationship between deposition and ANC. In order to estimate the amount of NOx and SOx deposition that will maintain an ANC level above a given limit requires the knowledge of the average catchment flux of base cation from weathering of soils and bedrock (i.e., preindustrial cation flux (BC0)). How might we generalize from location specific inputs (F-factor approach) to using this approach on a broader scale - watershed, regionally, or some other way - to generalize beyond individual locations? What other methods should be examined for estimating catchment weathering rates nationwide for surface acidity?

This is a challenging question and a difficult but critical problem. More attention is needed on the contribution of weathering rates – especially the release of base cations from soil primary minerals in affecting the recovery of terrestrial and aquatic ecosystems from acidification. The determination of base cation supply rates is critical to predict the effects of sulfur and nitrogen deposition on rates of acidification and possible recovery from acidification. Unfortunately there are no effective ways to estimate base cation weathering rates over large regions. Clarification is also needed on the utilization of the Bc (sum of base cations)/Al versus the more commonly used Ca/Al ratio in evaluating effects of soil acidity. Also, discussion is needed as to how the F-factor is applied in predicting the spatial and temporal patterns of base cation supplies and resultant effects on acidity.

It is vital to separate the capacity effects from intensity effects in assessing sensitivity to aquatic and terrestrial ecosystems to acidification and the potential recovery from acidification. An acidic soil is a necessary but not sufficient condition for the acidification of waters – a second requirement for acidification of waters is the introduction of mobile strong acid anions, such as sulfate and nitrate. Soils that are already naturally acidic will produce acidic waters nearly instantaneously with the introduction of mobile strong acid anions, and conversely, acidic waters in these cases will recover almost instantaneously when such mobile strong acid anions are removed. On the other hand, acidification of a soil that was not historically acidic is not so easily reversed.

In the consideration of impacts on ecosystem services, acidification effects are placed in a broader context. The section on ecosystem services is a good summary of helpful information related to ecosystem services and acidification issues. It is useful that the REA explicitly states some of the issues in estimating directly how ecosystem services are affected by terrestrial and aquatic acidification.

3. Section 4.3 and Appendix Y describe the analyses used to evaluate the effect of terrestrial acidification. This analysis uses the Simple Mass Balance Model to determine the impact of current deposition levels on Bc/Al levels relative to three potential Bc/Al cutoff levels (0.6, 1.2, and 10.0) for sugar maple in the Kane Experimental Forest and red spruce in the Hubbard Brook Experimental Forest and a larger assessment area based on the FIA database for 17 states. Is this approach adequate to develop critical loads of deposition for the broader terrestrial acidification case study area? Is the regression analysis between

Bc/Al ratios and tree health sufficiently described and are uncertainties adequately characterized?

The extrapolation of the critical load calculations for sugar maple and red spruce to other regions (e.g., different states) beyond those of the case studies helps place these results in a broader geographical context. It is not clear, however, whether the approach will be adequate for predicting effects in all sensitive areas. It is a surprising result that such a high percentage of sites have been compromised by acidifying total nitrogen and sulfur deposition in 2002 (page 4-62). The limitations of the USA FIA data in analyzing critical loads to high and medium elevation forests in the northeastern US needs further evaluation.

In this section there is some use of the use of average critical loads related to three levels of protection. It is not clear if this "average" is meaningful in the context of critical load determinations due to critical loads being dependent on the specific edaphic features of each area. Does the average critical load take into account the spatial distribution of specific edaphic features? Does this "average" apply to specific case study areas or to larger regions? The discussion and analyses that show how specific factors, including parent material and soil properties, affect critical load calculations need to be developed further due to the importance of these factors in affecting acidification of both terrestrial and aquatic ecosystems. How does variation in these edaphic factors both within and among regions affect the uncertainties in critical load calculations?

Nutrient Enrichment:

The title of the chapter was a point of discussion amongst the Panel. Some found "Nutrient Excess" to be a more accurate because nutrient enrichment commonly occurs but does not necessarily result in an environmental problem unless the ecosystem is not able to fully assimilate nitrogen inputs. Further background information should be provided on nitrogen as a limiting nutrient in terrestrial, freshwater aquatic and coastal ecosystems. Most temperate terrestrial ecosystems are nitrogen deficient and increased deposition may cause increased growth, which can be viewed as positive or negative. Recent evidence suggests that nitrogen limitation is more common than once thought for freshwater ecosystems. A major challenge in developing protocols for returning ecosystems to a level of lower nutrient enrichment is defining what attributes and their specific values are necessary for the restoration of ecosystem structure and function. The attributes that control sensitivity of different ecosystem types to nitrogen loading needs further clarification. It would be helpful to indicate the spatial extent of those areas in the West (e.g., Rocky Mountains of Colorado) that are impacted by relatively low levels of atmospheric nitrogen inputs.

1. Section 5.2 and Appendix 6 describe the analyses used to evaluate the effect of aquatic nutrient enrichment. The analysis uses the SPARROW model on one stream reach (Potomac River and Neuse River) to determine the impact of atmospheric total nitrogen deposition on the eutrophication index for the estuary. Does the Panel think that the model is adequately described and appropriately applied?

The SPARROW model and its links to the CMAQ model's deposition data and to the ASSETS EI's estimates of current or future occurrences of eutrophication are well described in the Appendices. However, better description of the SPARROW model is needed in the main document. EPA should bring forward some of the model's description in the Appendix 6 to improve readability of this chapter. Also, it would be helpful to clearly indicate that SPARROW is a statistically-based, steady-state model. The limitations of using this model for extrapolation to conditions of lower atmospheric nitrogen deposition should be given.

The Panel found the broader extrapolation of the SPARROW results helpful in discussion of the Potomac River/Potomac Estuary and Neuse River/Neuse River Estuary case studies. The discussion of the uncertainty estimates provides important information on the application of the model simulations. A summary should be provided to help the reader identify which component of uncertainty is most important with respect to policy recommendations

2. Section 5.3 and Appendix 7 describe the analyses used to evaluate the effect of terrestrial nutrient enrichment. This qualitative analysis describes the impacts due to nitrogen deposition on the Coastal Sage Scrub community in California and in mixed conifer forests in the San Bernardino and Sierra Nevada Mountains and larger areas where possible. In addition, the effects of nitrogen deposition in the Rocky Mountain National Park supplemental case study location are summarized. How would the Panel apply the threshold values presented in this case study to allow for a broader geographic application that accounts for regional variability? Have the associated uncertainties been adequately characterized?

Considering the general scarcity of ecological effects data due to excessive atmospheric N deposition, the use of a "patchwork quilt" of species and ecosystem types from across the United States is a reasonably acceptable option. The Panel finds it will be difficult to translate the results from these case studies to other ecosystems because the selected ecosystems represent regions that are ecologically sensitive to total reactive N deposition. Recognizing that a secondary standard must protect the most sensitive ecosystems, the Panel recommends that N-limited ecosystems (which are far more common than suggested by these cases) should be duly considered.

The emphasis on the California coastal sage scrub (CSS) and San Bernardino Mountains mixed conifer forest (MCF) systems seems appropriate due to their importance with respect to population centers. Also linkages with other environmental issues, such as fire susceptibility and the potential effects on biodiversity and threatened species, are important for these case study areas. The alpine ecosystems of the Rocky Mountains were also considered and the suggestion that these systems have "the ecological benchmarks ….comparable to the benchmarks from CSS and MCF ecosystems" (p. 5-57) needs to be reconsidered since these systems are especially sensitive to low levels of nitrogen deposition. The section on "Uncertainty and Variability" (5.3.8) is a good summary of the major issues with a particular focus on the CSS and MCF case studies. On the other hand the "Conclusions" (5.4) section should be expanded, linking the chapter with the entire document.

The main question remains – how to translate ecological changes caused by the total deposition of reactive nitrogen into ambient concentrations of a single criteria pollutant, NO_x? The critical loads approach based on deposition of total reactive N should be considered for setting a new secondary standard.

<u>*Additional Effects (Chapter 6):*</u>

1. *In this chapter, we have presented results from some qualitative analyses for additional effects including visibility, climate and materials, the interactions between sulfur and methylmercury production, nitrous oxide effects on climate, nitrogen addition effects on primary productivity and biogenic greenhouse gas fluxes, and phytotoxic effects on plants. 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?*

The Panel appreciated improvements to the chapter on additional effects of N and S deposition. Notwithstanding specific editorial comments and questions, the Panel concluded that the sections on visibility and materials, sulfur and mercury methylation, N_2O production, methane emissions, phytotoxic effects of gaseous NO_x and SO_x and productivity changes from N deposition had appropriate content and context for the REA. However, the discussion of N deposition effects on carbon cycle processes and carbon sequestration was found to be inconsistent with the available literature and EPA is requested to look at the specific comments provided by panel members to correct the existing text.

<u>*Synthesis of Case Studies (Chapter 7):*</u>

The synthesis chapter provides a summary of the disparate case studies, but does not fully synthesize their results. The chapter also does not provide the type of contextual information needed to extend these results to broader, national scales. Rather than reiterating the case studies' summaries, the Panel suggests revising the chapter to better reflect the sum of the parts and possible linkages among them. Potential foci include developing common category labels (comparable levels of concern for the different environmental effects), exploring how ecosystem services might be used as a way to focus and contrast the different case studies, developing illustrative figures that integrate across case studies, and adding an uncertainty section that synthesizes the net sum of and implications of the multiple uncertainties rather than just listing the individual components. In regard to the latter, the Panel suggests addressing the key uncertainties that have the most important policy implications, for example. The chapter should also include a section that identifies the major research and data gaps, including those that would allow spatial scaling of welfare effects and relationships between SO_x, NO_x and ecological structure and function.

1. *Here, the case study analyses are integrated and synthesized within the conceptual framework of ecosystem services as shown in Figure 7-2. Where possible, we have quantified select ecosystem services associated with the ecological effects targeted in this review. This chapter discusses adversity by characterizing the degree to which ecological effects are occurring under given levels of deposition to inform the discussion of adversity in the policy assessment and standard setting process. To what extent do you think the description of ecosystem services provides a useful framework in the case study*

analyses for informing standard setting? Does the Panel have suggestions for additional considerations or characterizations for ecosystem services relative to the case studies?

The concepts of ecosystem services are useful to help explain the implications of the effects of N/S deposition on ecosystems for public welfare, and support the policy assessment of what standards are needed to prevent adverse welfare effects. The Panel suggests EPA include a comprehensive description of ecosystem services expected to be affected by N/S deposition, and not limit the discussion to those services that can be quantified, such as recreation fishing or commercial timber production. What is most important is a description of how the abundance and quality of ecosystem services are affected by the currently observed effects of N/S deposition on ecosystems. Total services at current deposition levels do not reflect what has been lost due to current deposition effects, and only a portion of current services are at risk if deposition continues or increases.

2. *Based on the information presented in the current Risk and Exposure Assessment, given adequate time and resources, is there enough information to inform setting separate standards based on the other targeted ecological effects, specifically, terrestrial acidification, aquatic nutrient enrichment, and terrestrial nutrient enrichment? If not, how can our understanding of these ecological effects be enhanced in time to inform the next 5-yr review?*

Based on the Panel's understanding of the Agency's court-ordered deadlines for completing this review, we have recommended a hybridized approach whereby the EPA can fulfill its statutory obligations and move forward with an expedited review to set an ecologically relevant secondary NAAQS. With adequate time and resources to further develop the approaches being considered and identify the needed foundational analyses, the Panel finds the information in the current REA sufficient to set separate standards for terrestrial acidification, eutrophication of western alpine lakes and terrestrial nutrient enrichment. However, the Panel believes that setting a standard for coastal nutrient enrichment would be difficult because of the substantial inputs of non-atmospheric sources of N to these systems. Further research on cause and effect relationships driven by acidification and excess nutrient enrichment will facilitate future rulemaking.

Enclosure C

Compilation of Individual Panel Member Comments on EPA's *Risk and Exposure Assessment for Review of the Secondary National Ambient Air Quality Standards for Oxides of Nitrogen and Oxides of Sulfur: Second Draft (EPA-452/P-09-004a)*

Dr. Praveen Amar

First of all, this second draft of the REA "Risk and Exposure Assessment for Review of the Secondary National Ambient Air Quality Standards for Oxides of Nitrogen and Oxides of Sulfur" is a substantial improvement over the first REA draft. It is very well-written, is more complete, emphasizes important issues of risk and exposure at the right level, and is organized and structured in a manner that makes it much more readable.

In addition to general comments on the section of REA dealing with air quality analysis (Chapter 3, "Sources, Ambient Concentrations, and Deposition"), detailed comments are also provided on the Executive Summary.

<u>Comments on Executive Summary</u>

The charge question relating to the Executive Summary notes:

"In response to the Panel's review of the first draft REA, we have included an executive summary of this document. Does the Executive Summary adequately summarize and characterize the key issues driving this review as well as the important findings of the analysis? Does the Panel have any suggestions for clarification or refinement of the Executive Summary? "

I think it is very important to include an Executive Summary for this REA. At 24-page length, it is really more of a "synthesis report/document" than an "Executive" summary and that is good. My general impression is that, as written, it presents a good synthesis of all the *issues* raised in the REA. However, I believe that it needs a better focus on the summary of the *findings* described in various Chapters (Chapters 3,4,5,6, and 7).

The eight policy-relevant questions (page ES-3) do provide a focus on what needs to be done in the next phase of this CASAC review process. What, however, is not clear to me, is how these questions actually will be answered in the next "policy phase" of this process. Should EPA have made the first attempt to answer these questions here in this REA? What is missing in the Executive Summary and in the rest of the REA is a clear roadmap (or, a serious attempt) of how REA will get to the next "policy assessment" phase.

The articulation of the "right questions to ask" right at the beginning of the REA process is an excellent idea, even though it is difficult to formulate and write them out clearly. It is not clear to me why the List of policy-relevant questions in Chapter 1 (Section 1.4) is expanded to fifteen questions from original eight questions in the Executive Summary. Some of the questions in Chapter 1(Section 1.4) are included in the Executive Summary, but new ones are also added. I recommend that both Lists include the *same* set of policy-relevant questions. If the two Lists which follow each other need to be different, please do provide a rationale. Otherwise, it is confusing to observe that the List of questions is almost doubled right in the beginning of REA. If one assumes that the List of eight questions in the Executive Summary is more "focused" and more 'Executive" in nature, then the sequence of questions in Chapter 1 is not at all "parallel" to what is in the Executive Summary. For example, Questions 1, 2, 3, 4, 5, 6, 7, and 8 in the Executive Summary are Questions 6, 7, 2, 4, 8, 12, 14, and 15. This also means that Questions 1, 3, 5, 9, 10, 11, and 13 in Section 1.4 are not mentioned at all in the Executive Summary.

Also, the sequence of policy-relevant questions should follow the layout of the REA itself. For example, the structure shown in Figure ES-2 (air quality indicators to deposition metric to ecological indicators to establishing standards to determination of whether these standards are met) is a good way to re-arrange the sequence of policy-relevant questions.

The first and second questions in Executive Summary need to clearly say "to what extent do the current SECONDARY standards provide protection…" and "to what extent does the current SECONDARY NOx standard provide protection…." The fifth policy-relevant question (ES-4) should be modified to also include the corollary and related and important policy-relevant question "Does the available information provide support for considering *joint* air quality indicators for NOx and SOx?" The third policy question needs to address uncertainty in addition to variability "associated with those responses." Also, fifth bullet on Page 1-17 is awkward ("… trying to be protected against?").

A general comment about reduced nitrogen/NH_3/NH_4, etc.: This REA is a substantial improvement over the ISA and previous REA (first draft) in how it addresses the role of ammonia and reduced forms of nitrogen in total nitrogen deposition (however, it still does not address organic forms of nitrogen). The EPA staff deserves to be commended for this major shift in their treatment of total reactive nitrogen. However, there are references in the Executive Summary and in the body of the document (Chapter 3 and other places, see below) that ammonia is a "local" pollutant or that the ammonia sources are located in "rural" areas or that at four million tons per year, U.S. emissions are "small," compared to emissions of NOx and SO_2. These statements seem to imply that ammonia is not as important as the other two pollutants.

Recent work by EPA (Dr. Robin Dennis and others), however, indicates that even local "hot spot" emission sources (such as a CAFO or confined animal feeding operations) have a large regional total deposition footprint in addition to a more than minor total deposition local footprint. For example, they state that "the range of influence of a concentrated source of NH_3 (high-emission cell) is 200-400 km; 300 + km may be a good estimate." Since there is a large regional component associated with emissions, atmospheric chemistry, and transport of the NOx-SO_2-NH_3 system in addition to important "local" component, it is important that this REA, including the Executive Summary, be revised appropriately to better reflect the regional nature of ammonia emissions, and the regional role of reduced nitrogen in deposition and in ecological effects. The results presented in Chapter 3 clearly show reduced nitrogen deposition is from 20 percent to as high as 50 percent (above 30 percent on average) of total nitrogen deposition at locations of various case studies. Therefore, ammonia/reduced nitrogen needs to be treated the same way in the REA as SOx and NOx.

The Atmospheric Deposition Transformation Function (ADTF) and Ecological Effect Function (EEF) as shown in Figure ES-2 and as described in the Text are good conceptual framework to address "source to welfare effect". What is missing here, however, is an acknowledgement that these functions are simply not some "magic translators" but are very difficult to "derive," once

EPA takes into account geographical and seasonal variability of the relationship between concentrations and deposition, as well as uncertainty associated with measurements and model predictions. This needs to be clearly addressed in the context of Figure ES-2 and at other places where this Figure appears as the organizing principal of this REA.

Page ES-6: The Executive Summary and the document use “nitrogen enrichment,” (please see Page ES-6), “nutrient enrichment,” (see Table on Page ES-7) and “nitrogen nutrient enrichment” to mean the same thing. Please choose one (I suggest “nitrogen nutrient enrichment”) and use it throughout the document for clarity and for ease in reading.

Page ES-8 (line 6): “broad look” needs to be replaced with something more rigorous. For example, “broad evaluation/investigation” etc. The description on Page ES-9 (top) of monetized and non-monetized benefits is excellent.

Page ES-11: Figure ES-5, Title should say, NOx, and not, NOy.

Page ES-13: As an extension of my comment about reduced nitrogen/NH_3 above, it would be very helpful that the Executive Summary include two additional Figures in addition to the Figure ES-7, one on spatial distribution of oxidized nitrogen and the other of reduced nitrogen, with accompanying Text that highlights the important role that reduced nitrogen plays in conjunction with the role of oxidized nitrogen.

Page ES-15: It is important that this Executive Summary describe how the case studies will be “extrapolated” from “smaller scale to other sensitive areas in the country.” This important concept has been described in previous EPA efforts and needs to be addressed here before the four main ecosystem effects are presented

Page ES-19: Aquatic Nutrient Enrichment: The Text states “human activity has likely contributed to a six-fold increase in nitrogen flux ….” Note that REA (Chapter 3) notes that this has happened over the last 100 years. I would speculate that this large increase has occurred over a much shorter time horizon, of say, last 30 to 40 years, over which emissions of NOx from some industrial sources, and especially, ammonia from animal operations have substantially increased at an accelerated pace. If this could be confirmed by EPA, it should be noted in the REA, because this finding may be policy-relevant if it implies a larger and more recent role of reduced nitrogen in total nitrogen deposition.

Page ES-20: The ASSETS index needs to be spelled out and clearly explained here as well under “Key Terms,” especially under “Key Terms” in its proper context. I found it hard to understand. For example, it should be clear under “Key Terms” that it applies to nitrogen enrichment. Please also explain the use of the term “pressure” in its proper context. Same for “Determined Future Outlook” for the ASSETS index. The term, ASSETS itself, needs to be spelled out.

In “Key Terms” and in other sections including Executive Summary, the definition of “Uncertainty” needs to recognize that there is more to uncertainty than simply “parameter uncertainty.” Please note that in addition to parameter uncertainty, there is equally important, if not more important, the concept of model uncertainty (that sometimes is addressed through expert solicitation or other methods). It would also be very helpful to explain here that uncertainty is different than other terms with which it gets confused (for example, variability, precision, sensitivity, and risk). It would be also useful to explain all these other concepts under "Key Terms."

Page ES-22; The various ecological thresholds for CSS community need to be summarized. This is too much detail for the Ex. Summary.

Page ES-24, Conclusions: This section needs to be redone and rewritten. It does not do justice to an otherwise well-written Executive Summary. The words "confidence," "most confidence," "fair amount of confidence" may be following the recent IPCC format, but they just do not fit here. One suggestion is to write this Conclusion section in the excellent format of "Key Findings," prepared by EPA staff during the earlier phase of the REA process.

Comments on Chapter 3

The **first** key charge question relating to air quality analyses deals with treatment of uncertainties associated with air quality analyses in Chapter 3 and whether uncertainties have been identified and described appropriately. Chapter 3 describes EPA's chosen approach that characterizes the spatial and temporal patterns of deposition at locations of chosen case studies. The deposition is characterized for both wet and dry forms and for total reactive nitrogen (and its two components of oxidized and reduced forms) as well as for total sulfur.

The discussion of "uncertainties" in Section 3-5 is descriptive in nature. It is, however, not helpful in providing a quantitative and relative sense of uncertainties in various components (emissions, wet and dry deposition, sulfur versus nitrogen deposition). For example, it would be useful to state that NH_3 emissions are much more uncertain than NOx emissions that in turn are less certain than SO_2 emissions. It may also be useful to state that dry deposition is simply not measured but inferred from model calculations and has therefore uncertainties of unknown magnitude. For such cases, sensitivity studies could be useful to put bounds on the results. "Uncertainty" in "Key Terms" also needs to recognize that there is more to uncertainty than simply "parameter uncertainty" (see above).

The s**econd** charge question on air quality analyses is related to the replacement of Response Surface Modeling (RSM) in the first draft of REA with a new series of full-blown CMAQ simulations. The REA now contains CMAQ simulations to estimate the relative contributions of emissions of NOx and ammonia to nitrogen deposition (total N, and its oxidized and reduced forms) and a similar analysis for contribution of SO_2 emissions to total sulfate deposition. The second charge question asks, "Does this approach enable us to adequately examine the contribution of NOx to total nitrogen deposition?"

The replacement of RSM model with CMAQ and associated sensitivity runs (impact of 50% reduction in NOx, NH_3, and SO_2 emissions on deposition of nitrogen and sulfur) is step in the right direction. Evaluating the inter-annual variability in emissions and meteorology and their impact on deposition fluxes is an important and useful addition to this REA. The results in Chapter 3 and Appendix 1 are described in great detail in various figures, tables, and charts. My major comment, however, is that a much better and more realistic approach to investigate the impact of emission reductions on deposition fluxes is through the application of " Direct decoupled Method"(DDM) than the "brute force" approach of reducing emissions of one of three pollutants by a set percentage (in this case, 50%). Realistically, emission strategies are not based on "one pollutant at a time" reduction, but are a combination of different reductions in the emissions of SO_2, NOx, and NH_3. The relative sensitivity of various components of deposition fluxes (wet, dry, total, oxidized and reduced nitrogen, sulfur, etc.) should be investigated more meaningfully through the application of DDM or similar approach. It is not clear if EPA would be able to undertake such an effort in the short time available to develop secondary standards for SOx and NOx.

My second major comment (also noted above in the Executive Summary and repeated here) is that this Chapter needs to more clearly address ammonia and its emissions and impacts on deposition. Statements such as ammonia emissions are "local" or "rural" or are not as widespread as NOx and SO_2 miss the important point that ammonia is an extremely important chemical in the context of establishing secondary standards for SOx and NOx. Even though I do recognize that under the current regulatory constraints, REA can only develop a secondary NOx standard with ammonia "embedded" in it, the task is going to get only more difficult if we do not address the issue of ammonia and reduced nitrogen clearly.

The **third** charge question on air quality analyses relates to the CMAQ model performance evaluation undertaken by EPA after the first draft of REA in response to recommendations made by our Panel. This charge question asks, "Is this (new) analysis sufficient to support the use of the model in this review?"

Appendix 1 does provide useful and clearly presented information on the evaluation of CMAQ for annual average concentrations and annual wet deposition fluxes for components of nitrogen (nitrate, ammonium, sulfate, etc.) for the year 2002. It also provides data on model performance by comparing predictions of monthly concentrations and wet deposition fluxes with measurements for the four-year period of 2002-2005. Though model-measurement comparisons are useful for annual and monthly time scales, it may be useful to evaluate model at finer temporal scale (for example, hourly and weekly data where available) for a more "stressed" performance evaluation of CMAQ's results.

Dr. Andrzej Bytnerowicz

Executive Summary

Does the Executive Summary adequately summarize and characterize the key issues driving this review as well as the important findings of the analyses?

Generally, it does. The section is well written and provides a good overview of the entire document. Graphics, maps and tables are informative and useful. However, some corrections are needed (see my replies to the next question).

Does the Panel have any suggestions for clarification or refinement of the Executive Summary?

a. the second paragraph in the box of page ES-1 should be corrected to: "Substances known as oxides of sulfur, or SOx, include multiple gaseous substances (e.g., sulfur dioxide [SO_2], …."

b. page ES-2 – in the end of the last paragraph a statement that biodiversity changes have also been observed in other ecosystems, such as coastal sage scrub, mixed conifer forests in California or alpine ecosystems of the Rocky Mountains, should be added.

c. page ES-11, text and Figure ES 5 – while the text described NOx, the figure shows NOy. Consistency is needed here. On the same page, line 3, change "Peak SO_2 concentrations" to "Highest annual SO_2 average concentrations".

d. page ES-18, lines 13-16 - this paragraph seems to be out of place. Consider deleting.

e. pages ES-21 through ES-23, section "Terrestrial Nutrient Enrichment" – references should be added to this section.

f. page ES-23, Table ES-4 – zeros for N deposition > 17 kg N/ha/yr do not make sense. Please change to some meaningful values.

Case Study Analyses

1. Are uncertainties appropriately characterized across the case studies? Is there adequate information to allow us to weigh the relative strengths of each case study to inform the standard setting process?

Yes. For all described cases (acidification of aquatic and terrestrial ecosystems; nutrient enrichment of aquatic and terrestrial ecosystems) uncertainties have been well described and examples of potential problems have been given. A good example is a statement on page 5-41 describing uncertainties related to the SPARROW model: estimates of N loading based on the CMAQ/NADP overall N loading in the model calibrated against the wet nitrate deposition values. Similarly, for the nutrient enrichment case studies for the terrestrial ecosystems, problems and uncertainties related to air pollution monitoring techniques (such as for ammonia), those due to limitations in the spatial extent and density of monitoring networks, or uncertainties due to the design and performance of the CMAQ model are listed.

2. In using the risk and Exposure Assessment to inform the policy assessment, we plan to focus on aquatic acidification as the basis for an alternative multi-pollutant secondary standard as this is the area where we have the most confidence in our ability to characterize adverse effects. Does the Panel agree with this approach?

Yes. This research is most advanced and well documented and therefore I agree with such a selection.

<u>Nutrient Enrichment</u>

1. Section 5.2 and Appendix 6 describe the analyses to evaluate the effect of aquatic nutrient enrichment. The analysis uses the SPARROW model on the stream Reach to determine the impact of atmospheric total nitrogen deposition on the eutrophication index for the estuary. Does the Panel think that the model is adequately described and appropriately applied?

The SPARROW model and its links to the CMAQ model that provides deposition data and to the ASSETS EI that estimates a likelihood of the current or future occurrence of eutrophication are well described in the Appendices. A better description of the SPARROW model is needed in the main document. Therefore I suggest that most of the information contained on pages 6-31 through 6-33 and Figure 2.2-3 of the Appendices is copied into Chapter 5 of the main document. Otherwise a reader has to read both volumes to understand the SPARROW model and the entire methodological approach.

2. Section 5.3 and Appendix 7 describe the analyses used to evaluate the effect of terrestrial nutrient enrichment. This qualitative analysis describes the impacts due to nitrogen deposition on the coastal sage scrub community in California and in mixed conifer forests in the San Bernardino and Sierra Nevada Mountains and larger areas where possible. In addition, the effects of nitrogen deposition in the Rocky Mountain National Park supplemental case study location are summarized. How would the Panel apply the threshold values presented in this case study to allow for a broader geographic application that accounts for regional variability? Have the associated uncertainties been adequately characterized?

The proposed approach of an assemblage of a “patchwork quilt” of species and ecosystem types from across the United States is a reasonable and probably the best option considering a general scarcity of data on the ecological effects of atmospheric N deposition. Research results from the ecosystems selected for the case study (coastal sage and mixed conifer forests), as well as from alpine ecosystems in the Rocky Mountains, north-eastern forests, and mid-western grasslands, provide a reasonable representation of the continental US ecological zones, and a high probability that comparable responses would take place in other zones. A range of benchmarks is based on the well characterized sensitive ecological indicators such as changes in lichen communities, especially decrease of nitrophytes at the low end of the sensitivity spectrum; changes in mycorrhizal associations in the mid-range of the spectrum; and nitrate leaching to streams at high levels of atmospheric N deposition. Changes in lichen communities seem to be the most promising N deposition indicators of N deposition effects which could be used nationwide - comparable changes of epiphytic lichens in various ecological zones can be expected at the similar levels of N deposition since such lichens get their nutritional N from the atmosphere. However, when considering responses of lichens to N deposition, also other factors such as phytotoxic ozone or climatic differences have to be taken into account. Changes in higher plants used as sensitive indicators of N deposition would be characterized by much larger margins of uncertainty because of the edaphic, climatic and many other differences in ecosystems or ecological zones in the US.

The main question still remains – how to translate changes caused by the total nitrogen deposition just into the ambient concentrations of the criteria pollutant (i.e., NOx). Since ecological indicators included in this analysis, as well as the biological systems in general, react to the total reactive N (including its reduced inorganic and organic forms), this seems to be the most important problem to solve.

Additional problem is related to the N deposition data produced by the CMAQ model. Improvements of the model are needed in order to include N organic species in total N deposition estimates as well as the more recent, finer resolution data.

<u>Other Comments</u>

Page 5-30, Figure 5.2-12 – please check the legend. Number of a group should be assigned to each color. Number of estuaries is already listed in Table 5.2-2 and should not be duplicated in the table.

Page 5-32, line 6 – change "reductions" to "decrease".

Page 5-34, Figure 5.2-15 – drawing a line through these data points is highly problematic.

Page 5-55, lines 3-8 – there is also a need for improving the dry deposition methodologies through more robust models aided by empirical, ground level estimates based on well developed monitoring networks.

Page 5-66, line 11 – in contrast to fire suppression policy and increased atmospheric N deposition, ozone does not contribute to increasing stand density.

Page 5-72, line 3 – add "of exotic species" after "invasion".

Page 5-79, line 11 – check the units.

Page 5-81, line 20 – add "especially in remote areas" after "networks".

Page 5-85, lines 10-12 – reference has already been cited above.

Pages 6-29 through 6-42 – check references, many cited in the text are missing.

Ms. Lauraine Chestnut

Final comments on Risk and Exposure Assessment for NOx/SOx secondary standard review, August 5, 2009

Executive Summary

Introduction: Setting the stage for this assessment would benefit from a short paragraph explaining why this secondary standard review is focusing on the ecological effects of NO_x & SO_x deposition, and not covering other potential welfare effects such as foliar injury from gaseous phases of NO_x & SO_x or other effects of deposition such as injury to materials.

Page ES-2: The end of this section gives a description of the extent of the problem (a description of the effect and its geographic extent) for the effects of N deposition through excess nutrient enrichment in terrestrial ecosystems. Similar descriptions should be added for acidification and eutrophication. For example, the discussion on page 4-9, lines 18-28, does this well for acidification of aquatic ecosystems in the US.

Policy-relevant questions: The document does not appear to attempt to directly answer these question. Is this something that is going to be done in the policy document? It would help the reader to understand where to expect to see these answered. The first question seems very important, although perhaps a bit too broad. This review is looking at only ecosystem-related welfare effects associated with NO_x & SO_x deposition. When will the case be made that current standards for SO_2, NO_2, $PM_{2.5}$ and ozone (of which the latter two are currently exceeded in many locations) are not sufficient to remedy or prevent ecosystem-related welfare effects from NO_x & SO_x deposition?

Page ES-5, lines 18-20: The relationship between the standard and the "maximum deposition load" still seems a little vague. The standard would be the maximum ambient concentration (in the air) that would keep the deposition level at or below the maximum deposition load? The maximum deposition load is determined based on maintaining the ecological indicator at or below the level determined to be acceptable in the policy assessment? Please clarify the use of this term in this document versus the term "critical load," which seems to have a somewhat different usage in the scientific literature.

Figure ES-2: The logical flow of this figure is good, but I still have questions about the spatial dimension. Seems like this type of standard would allow ambient concentrations to be higher in locations where it does not lead to unacceptable levels of ecosystem indicators, which may make sense but is an unusual approach for a NAAQS. The geographic scale on which the standard will be assessed as met or not will have to be addressed somehow.

Page ES-6, line 10: Insert "selected" in front of "ecosystems", and add some explanation of why these ecosystems were selected as case studies. They aren't the only sensitive ecosystems—do they have the best data, are they most representative, are they the most sensitive? Any way to give a sense of what share of the problem in the US that these case studies represent for each of the four categories of effects?

Page ES-8: It is good to be working in the concepts of ecosystem services and how that relates to the CAA definition of welfare effect. Be careful to include nonuse values such as habitat preservation in the descriptions of ecosystem services because there are many aspects of ecosystem services that are important to the public even though they do not involve direct human use. For example, I benefit from knowing that lakes in the Adirondacks support aquatic life without significant loss of quality due to manmade emissions even though I never intend to go fishing there. This type of ecosystem service is mentioned several places in the assessment, but is not mentioned in the sections on aquatic acidification. Even though the only service intended for quantification is recreational fishing, nonuse types of services should also be listed and described.

Page ES-12: Add some sentences on the key conclusions from chapter 3 about the analysis of oxidized and reduced N.

Page ES-13: In what locations are acidification effects a problem in the US? It is not the whole country.

Page ES-18, lines 13-16: Some data are presented on forest recreation usage. It would be good to make a connection between these activities and the effects of acidification. Presumably there is some degradation in the quality of experience, and perhaps even loss of area suitable for some types of recreation, due to declining forest health. Similar information for aquatic acidification could also be mentioned, as it is included in Chapter 4. It is important to note both here and in Chapters 4 and 5 that total ecosystem services at current deposition levels do not reflect what has been lost due to current deposition effects, and only a portion of current services are at risk if deposition continues or increases. Some connections between the quantity and quality of these ecosystem services and the effects of NO_x & SO_x deposition need to be made, even if the connections are only descriptive.

Page ES-19: How significant and extensive is the problem of eutrophication in US coastal estuaries?

Page ES-20: It is a very significant finding that more than 100% reduction in NO_x deposition would be needed to move the case study areas from bad to poor on the indicator scale. How generalizable is this conclusion? Does NO_x cause a comparable share of total N deposition in other sensitive locations? Is this because total N deposition is a small share of the total nitrogen entering the estuaries? Be careful with the wording here. In Chapter 7 there is mention of a “weak” relationship between aquatic nitrogen and the indicator. The weakness is in terms of the effect of changes in NO_x deposition on the indicator, I think, not necessarily when all sources of N are considered. Also, be careful to avoid implying that there is no benefit of reducing NO_x deposition—it may not alone be enough to solve the problem, but it might be useful if part of a broader policy to reduce all N emissions.

Page ES-22: Some explanation of the relationship between lichen and forest health would be helpful here.

Page ES-24, line 14: Restrict this statement to say there is less confidence in the relationship with NO_x deposition, not with N enrichment as a whole. Are there any specific circumstances with these case studies that contribute to this result, or would all coastal estuaries be similar?

Detailed comments:

Page ES-2, line 11: Sounds like sulfur can lead to nutrient enrichment and eutrophication. May need to make two sentences here.

Page ES-2, line 16: replace “alters” with “can alter”

Page ES-6, line 10: Insert “selected” before “ecosystems”

Page ES-9, line 6: Line begins “remain unidentified”, should say “remain unquantified”

Page ES-16, line 20: Connect these effects categories to the ANC levels, as is done in Chapter 4.

Page ES-16, line 35: Insert a sentence on the results for lakes at various ANC thresholds as line 37 does for the streams.

Page ES-17, lines 14-15: What does reduction in fine root growth mean for tree mortality, growth, or susceptibility? It says more in Chapter 4.

Chapter 4, page 4-8

The current value of recreational fishing has presumably been lowered because of current effect of aquatic acidification, by lowering the number of lake with fish and lowering the number of fish in other lakes. In the absence of these effects there would presumably be more days spent in this activity and many days would have better quality (with more locations available and better fishing experience at locations now being used). The total value today does not say much about what the change in value would be (although it gives a sense of scale) if the effects of deposition were eliminated. Are there plans to make a quantitative link? Perhaps this is part of the policy assessment?

Dr. Ellis B. Cowling

In preparation for the July 22-23, 2009 CASAC meeting, my individual comments on the Second 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 part in response to Charge Questions posed in Lydia Wegman's memoranda to Kyndall Barry dated June 5, 2009. As you will see, greater attention has been given to the Executive Summary and the several Case Study Analyses which were the specific assignments given to me by Chairman Ted Russell.

With regard to the Executive Summary, the principal Charge Questions were in essence:

1) **Does the Executive Summary adequately summarize and characterize the key issues as well as the important findings of this REA analysis?**
2) **Does the Panel have any suggestions for improvement of the Executive Summary?**

My general response is that it is highly desirable to have an Executive Summary in this Second Draft REA. My specific response to question 1, however, is that the Key Issues are presented very well but that the Important Findings are not so adequately presented. Thus, my suggestion for improvement of the Executive Summary is to give much more attention to the Important Findings (such as those that are presented in the "Summary of Case Study Analysis Findings" on pages 3-79 through 3-81 in Chapter 3, "Results for the Case Study Areas on pages 4-56 through 4-71 in Chapter 4, "Current Conditions in the Case Study Areas" on pages 5-18 through 5-43 and on pages 5-58 through 5-81 in Chapter 5, "Nitrogen Addition Effects on Primary Productivity and Biogenic Greenhouse Gas Fluxes on pages 6-13 through 6-28 in Chapter 6, and in the "Synthesis and Integration of Case Study Results" on pages 7-1 through 7-24 and especially the "Conclusions" section on pages 7-24 and 7-25 of Chapter 7.

In my opinion, the Important Findings in this Second Draft REA should be presented in the Executive Summary in the form of as direct answers as possible to each of the eight Policy-Relevant Questions presented for the first time on pages ES-3 and ES-4. It also would be desirable for these Important Findings to be presented in close physical proximity within the text of the Executive Summary to the presentation of Policy Relevant Questions to which these Findings/Answers to Policy Relevant Questions apply.

General Comments on the Chemical Forms of Total Reactive Nitrogen

It was a pleasure to see that both the Executive Summary and all of the main chapters (1 though 7) of this Second Draft REA give more appropriately balanced attention than was given in the First Draft REA to the **chemically reduced** as well as the **chemically oxidized** forms of the **inorganic parts** of total reactive nitrogen (Nr). But both the Executive Summary and each of the main chapters of this Second Draft REA do not give appropriate attention to the **organic** as well as the **inorganic** forms of **total reactive nitrogen**.

I was pleased to see that the term "Total Reactive Nitrogen" is listed among the "Key Terms" listed on pages xxi-xxviii of this Second Draft REA. But I was disappointed to see that the definition given for "total reactive nitrogen" in the "Key Terms" list is:

1) Not consistent with the way this same term is used in Chapters 3, 4, and 5, and also
2) Not consistent with the generally accepted scientific definition of this very important term.

In this connection, please note the perhaps subtle but very important differences between the definition of "total reactive nitrogen" as presented in the "Key Terms" list on page xxvii of this REA:

> "Total Reactive Nitrogen: All biologically, chemically, and radiatively active nitrogen compounds in the atmosphere and the biosphere, such as ammonia gas (NH_3), ammonium ion (NH_4^+), nitric oxide (NO), reduced nitrite (NO_2), nitric acid (HNO_3), N_2O, reduced nitrate (NO_3^-, and organic compounds (e.g., urea, amines, nucleic acids);" and the

definition of the term "reactive nitrogen" in the now publically available Executive Summary of the report of EPA's Science Advisory Board's Integrated Nitrogen Committee:

> "The term reactive nitrogen (Nr) is used in this report to include all biologically active, chemically reactive, and radiatively active nitrogen (N) compounds in the atmosphere and biosphere of Earth. Thus, Nr includes inorganic chemically reduced forms of N (NH_x) [e.g., ammonia (NH_3) and ammonium ion (NH_4^+)], inorganic chemically oxidized forms of N [e.g., nitrogen oxides (NO_x), nitric acid (HNO_3), nitrous oxide (N_2O), N_2O_5, HONO, peroxy acetyl compounds such as PAN, and nitrate ion (NO_3^-), as well as organic compounds (e.g., urea, amines, amino acids, and proteins), in contrast to non-reactive gaseous N_2."

In Chapters 3, 4, and 5, the terms "reactive nitrogen" and "total reactive nitrogen" are used to refer the sum of wet plus dry deposition of chemically reduced and chemically oxidized **inorganic** forms of air-borne biologically active nitrogen compounds and do not include the additional organic forms of airborne reactive nitrogen compounds (urea, amines, amino acids, and proteins).

In truth, at present, much more is known about the chemically reduced and chemically oxidized **inorganic** forms of reactive nitrogen deposited across the United States than about the amounts of organic forms of Nr deposited across the United States. But, at least in the Neuse River Estuary (which is one of the eight important "Case Study Areas" used in this REA report!), Hans Pearl has presented reliable evidence that as much as one third of the total atmospheric deposition of Nr compounds delivered to this estuary is deposited in the form of organic compounds -- in addition to the chemically reduced and chemically oxidized inorganic forms of Nr. Although it is not known how much organic nitrogen is present in the air sheds of the other Case Study Areas used in this REA, it would be appropriate to admit this uncertainty in the estimates of "total reactive nitrogen" and to use the term "total inorganic reactive nitrogen" or "total inorganic Nr" (for short) rather than the misleading "total reactive nitrogen."

It also would be desirable to include some discussion (or at least an acknowledgement) about the current uncertainty about the amounts of organic forms of Nr in the air-sheds of all of the Case Study Areas. This would be especially desirable in the various specific sections of this Second

Draft REA that deal with "Uncertainty" -- for example, on page 2-18 in Chapter 2, on pages 3-90 through 3-96 in Chapter 3, on pages 4-68 and 4-69 in Chapter 4, on pages 5-40 through 5-43 in Chapter 5, on pages 6-23 and 6-24 in Chapter 6, and on pages 7-21 through 7-24 in Chapter 7).

General Comments on the Major Ecological Effects of Reactive Nitrogen and Sulfur

It was a great pleasure to see very clear divisions and scientifically sound descriptions of the phenomena of excess Nr- and S- induced Acidification, Nutrient Enrichment, and Additional Effects in both terrestrial and aquatic ecosystems that are the object of the reviews of the ISA and REA documents we have reviewed so far in this integrated NOx and SOx NAAQS Secondary Standards Review process.

It is even more satisfying to see the very thoroughly thoughtout general approach to management of the ecological effects of excess Nr and S that is presented for the first time in Chapter 2 and is used as a "reminder symbol" before the title of all the main Chapters of this Second Draft REA document. I presume that the frequent use of this symbol is an indication of the pride that NCEA and OAQPS who collaborated in the creation of this general approach. Your pride in this accomplishment is well-deserved in both my personal and professional opinions.

The effort both organizations have made to develop this general approach – which is necessitated in part by the fact that chemically reduced forms of reactive nitrogen are not (yet?) recognized as Criteria Pollutants, and by the huge variety and complexity of the ecological phenomema that are of concern in the widely scattered geographical areas in which these phenomena are manifested – is especially commendable in light of the rather substantial departure from the dominating past concerns of the USEPA with largely urban- and suburban-based public-health rather than also public-welfare effects of air pollution in our country.

Permit me also to congratulate the authors of this Second Draft REA for their abundant and effective use of color illustrations that display many of the special features and unique challenges of management that will be necessary if the ecosystem protection approach that is necessary for success in establishing improved secondary standards for NOx and SOx as proposed in this REA.

It was particularly satisfying to see that the concept of Ecosystem Services has been so clearly explained and used in the development of this REA document. It also was very satisfying to see that the concept of Critical Loads is not only explained very properly but also used in presenting some of the recommendations in Chapters 4 and 7 in this Second Draft REA document.

Specific Continuing Concerns about Some of the Specific Words and Phrases Used in the Chapters of this REA

During the now nearly six years in which I have served as a statutory member of CASAC, I have offered persistent suggestions and recommendations to decrease the confusion that often results from the use of terms that have multiple meanings and thus frequently lead to lack of clarity in many of the ideas that are presented in the ISA, REA, and Staff Paper documents used in the NAAQS review processes.

The words and phrases that I once again call attention to in the context of this REA document include the following:

"**Level**" which most importantly is used in NAAQS review documents to mean "EPA designated allowable air concentration of a criteria pollutant." But the word "level" also is used to mean many other things such as:

1) "amount" of anything -- such as the amount of biomass lost, or amount of some chemical constituent in a water body,
2) "extent" of some physical or chemical phenomenon or even the number of people in a population that are concerned about some public health or public welfare impact of pollution,
3) "elevation" when altitude or distance above sea level is intended,
4) "degree" or "intensity" of some biological phenomenon or social concern,
5) "distance above zero" for example in a graph,
6) "Type of interest" such as at the "biological species level" as opposed to "physiological level" or "biochemical level," etc, etc.

In all NAAQS review documents, I recommend that the word "level" be reserved almost exclusively to discussions about the "EPA designated allowable air concentration of a criteria pollutant" and that the great variety of alternative words be used whenever they are in fact what is intended.

"Reduce, Reducing, and Reduction" – These three terms all have both chemical and numerical meanings. Fortunately we have the unambiguous terms "decrease" and "decreasing" which have only a single (always numerical) meaning.

Thus I recommend that the unambiguous term "decrease" be used instead of the word "reduce" when our intended meaning is numerical -- and thus reserve the term "reduce" exclusive for its chemical meaning? I further recommend that the couplet "chemically reduced" and "chemically oxidized" be used when referring to the two major chemical forms of inorganic reactive nitrogen.

A particularly interesting (and at the same time very frustrating!) example of an effort to follow this recommendation in this Second Draft REA document is provided by the wording for the captions of Figure 3.4-1, Figure 3.42-2, Figure 3.4-3, Figure 3.4-5, Figure 3.4-6, and Figure 3.4-7 on pages 3-82 through 3-85 in Chapter 3. Please note that the unambiguous word "decrease" was used in the captions of all six of these figures. For example, the **caption** for Figure 3.4-1 reads as follows:

> "Figure 3.4-1. The percentage impacts of a 50% **decrease** in NOx emissions on total reactive nitrogen deposition in the East." [The **Bolding** was added by me, for emphasis].

What a great disappointment it was to then read the description of this very same figure as it was written on lines 13 through 15 of the **text** on page 3-82 within Chapter 3!. The **text** reads as follows:

> "Figure 3.4-1shows the impacts of the 50% NOx scenario on total reactive nitrogen in the East. In general, a 50% **reduction** in NOx had a 30% to 40% impact (i.e., **reduction**) on total reactive nitrogen deposition. [Once again this **Bolding** was also added by me for emphasis.]

Please also note that **text** description of this figure refers (incorrectly) to the impacts of a "50% reduction in NOx" – which is not the same as a 50% reduction in NOx **emissions** -- as is stated (correctly) in the caption itself !!!

Nitrogen, NOx, NHx, Reactive Nitrogen, Total Reactive Nitrogen, and Total Inorganic Nitrogen . There are many examples in all seven chapters of this REA document where it is not clear whether the intended meaning of the sentence is best conveyed by the word "nitrogen," "NOx," "NHx,"" reactive nitrogen," "total reactive nitrogen," or "total inorganic nitrogen. I list below a few examples where the meaning of the sentence was were particularly puzzling:

1) Lines 13-20 on page 1-10 and lines 5-6 on page 1-19 in Chapter 1.
2) Lines 4-10 on page 2-6 in Chapter 2.
3) Lines 14-20 on page 3-3 in Chapter 3.
4) Line 6 on page 3-6 and line 13 on page 3-7 in Chapter 3.
5) Lines 5 and 6 on page 4-15 in Chapter 4.
6) Lines 5 and 6 on page 5-21 in Chapter 5.
7) Lines 27-31 on page 6-17 in Chapter 6.
8) Lines 7-9 on page 7-8 in Chapter 7.

Dr. Charles T. Driscoll

For each chapter general comments are given followed by specific comments.

Overall

The EPA is to be commended for this effort at compiling and completing the REA on ecosystem effects of sulfur and nitrogen oxides. This was a major undertaking and an important new initiative for future air quality management in the U.S. This report lays of a framework for a critical loads approach to guide mitigation of ecosystem effects from air pollution. The REA document is much improved over the previous draft. There are less typos and wording problems. Some of the redundancy has been eliminated. Most importantly the document is more focused than the previous draft

While I am enthusiastic about the initiative and report, there are a few considerations that should be addressed before the REA is finalized. First, there needs to be a more comprehensive and systematic discussion on the front end of the document on the concepts of maximum deposition load, critical load and target load. I have no idea what maximum deposition load is or how it is different from critical load. The term is introduced at the beginning of the report and then not really addressed again, while the term critical load is used throughout the report. Also the distinction needs to be made between critical loads and targets loads. This distinction is ignored in the report, but it has critically important policy considerations. Critical load is a steady-state concept. Unfortunately ecosystems are rarely, if ever at steady state. Steady-state models can be used to determine critical loads and/or empirical models. Target loads are a time –dependent phenomenon, and calculated with dynamic models. An important management consideration in this analysis is the time for ecosystems to reach critical chemical limits or achieve conditions of critical biological indicators following emission controls. Also the time to reach steady-state is important to understand for management considerations. The REA does not consider these time dependent process. The document really needs a clear treatment of these concepts and how they are used in the REA and for ecosystems management of air pollution effects.

Another important consideration is the spatial and temporal compatibility of the atmospheric transport models (i.e., CMAQ) and the watershed effects models. CMAQ is designed to address large spatial scales over short temporal scales. Many of the ecosystem effects and associated models are manifested over smaller spatial scales and over long time frames (e.g., decades to centuries). In the air quality chapter the authors provides some discussion of these considerations. If a critical load/target load is to be used to assess and guide ecosystem effects of air quality a rigorous analysis of the compatibility using transfers between atmospheric transport and watershed/ecosystem models this disconnect needs to be rigorously evaluated. While such an analysis is beyond the scope of the REA some text needs to be dedicated to this issue to set the stage for a future research initiative.

I have some pet peeves with the writing style that I have mentioned in previous reviews. Throughout the document the text is written as inanimate objects are doing something. For example, Figure xxx shows… This chapter discussed… An inanimate object cannot do anything.

A better approach would be: such and such is shown in Figure xx or In this chapter current emission sources are discussed…

I also urge that the word reduced only be used when referring to chemical reduction or a reduced chemical form such as ammonium. In the document the term reduced is used to refer to decreases as well as reduced chemical forms. This makes for some confusing text. Please make this change.

Executive Summary

General Comments

The quality of the Executive Summary is good. It provides a good overview of the report.

ES-2, line 10 - The text indicates that sulfur can limit productivity. I don't think this is a likely situation. The text should be clarified and probably corrected.

ES-5, line 18 – the REA uses a term maximum deposition load. How does this compare with the terms critical or target loads which are used later in the document (Chapter 4). Introducing a new term that is similar to other terms used in the report will confuse the reader. This term should be clarified relative to critical/target loads.

ES-16, text box – In this text box critical load is defined. I would like to see the actual definition of critical load, "the level of atmospheric deposition of a substance below which there is no harm to the ecosystem" or something like that. How does critical load compare with maximum deposition load? Also no effort is made to clarify the time-dependent nature of the problem. Critical load is a steady-state phenomenon; target load is a time dependent value. The text box should be expanded to address these issues.

Specific Comments

ES-2, line 7 – I do not like the term occult deposition. I do not believe it is a very accurate or descriptive term. Why not refer to it as cloud and fog deposition?

ES-2, line 13 – Change to … localized loss and extinction of fish.

ES-16, line 13- MAGIC should be defined or clarified.

ES-18, paragraph 2 – Some mention should be made of the value of sugar maple in fall foliage with respect to tourism.

Chapter 1.

General Comments

None

Specific Comments

1-13, line 4 – I do not like the term occult deposition. It is not a very accurate or descriptive term. Why not just refer to cloud and fog deposition?

1-13, line 6 - The text indicates that sulfur can sometimes limit plant growth and productivity. I don't think this is a likely situation. The text should be clarified and corrected.

1-15, line 21 – Methylation of mercury can occur in virtually all watersheds, not just the northeastern U.S and southeastern Canada. This sentence needs to be deleted or corrected.

Chapter 2.

General Comments

2-10, line 9 - As discussed above I do not like the term "maximum deposition load" it seems redundant with the concept of critical/target load. Why introduce a new term? This term should be clarified with respect to how it is different/ related to critical load. Also the second half of the phrase is not correct. It is not the amount that solves a mass balance equation for an ecological indicator. This sentence needs to be corrected.

Specific Comments

2-12. line 15 – The sentence "Valuation may be an important step…" is very similar to 2-11, line 18 "In addition valuation may be an important step…". You may want to delete one of these.

2-15, line 24- data are…

Chapter 3

General Comments

My hat is off to the authors of the chapter it is greatly improved over the last version. It is much more focused and easier to read. The authors did a great job re-orienting to focus on the ecosystem case studies. I also appreciate the effort made at the end of the chapter to discuss the sub-grid scale issues.

This section could be shortened further by eliminating the text on sulfur deposition for the nutrient case study areas; Potomac, Neuse, and western sites.

The authors should examine the text with respect the writing style. In some sentences the text is written as the present tense, in others it is written as the past tense. The text should be consistent, I would suggest in the past tense.

Specific Comments

3-1, line 21- Change to: were used as modeling…

3-2, line 8 – Change to: emissions, transformations and deposition…

3-2, line 9, 18- A typo? additional x? Should this be NHx?

3-2, line 28- Change to: The emissions to atmospheric concentrations-to-deposition...

3-3, line 17- Change to: are important components…

3-14, line 12- Change to: United States were also used.

3-15, line 16 – Aren't there about 200 NADP sites?

3-22, line 8 – The term non-ambient loadings is horrible. It doesn't mean anything. Please change it. I would suggest non-atmospheric N sources.

3-27, line 16- Should be Whiteface.

3-27, line 17- Change to: show a downward pattern to 2006.

3-48, line 8- Change to: is fairly uniform…

3-48 line 9- Change to: are generally consistent with…

3-59, line 10- Change to : Whiteface

3-80, line 10- Change to: and thus, decreasing atmospheric…

3-80, line 23- Change to: simulations were run.

3-81, line 23- space between to and 50%

3-82, line 5- Change to: for the pattern is that decreasing NOx decreases HNO3 which limits NO3.. There is a 4 before NO3, should this be HNO3?

3-82, line 5- Change to: This change… also NH4+ superscript on the charge.

3-82, line 8- Should this be dry deposition of NHx?

3-82, line 29- Should this be limits HNO3? Also a space between (NO3) and increases.

Chapter 4

General Comments

The acidification chapter is in need of some editing. Several sections are difficult to follow and the wording needs to be cleaned up. The term inorganic aluminum should be changed to dissolved inorganic aluminum, to clarify that the aluminum is in the aqueous phase not particulate phase. Throughout the chapter the authors refer to ANC concentrations. ANC is not really a concentration. It is a measure of acid base chemistry and represents the composite of many solute concentrations. It is possible to have negative ANC values; concentrations cannot be negative. The authors should refer to it as simply ANC or ANC value

4-14 In the section on critical loads, it is not at all clear what calculations are being done. This needs to be clarified. How are critical loads determined? Also critical load is a steady-state phenomenon, but model calculations were done for 2020 and 2050. Aren't these values really target loads? Finally critical loads are discussed here, but in the Executive summary the term maximum deposition load is introduced. How are these concepts different? In addition to the MAGIC calculations, the steady state water chemistry model (SSWC) is introduced on p 4-36. Both models should be introduced in this methods section. It should be made clear how the models are used and how they complement each other. On 4-36 and 37 the description of the SSWC model is very confusing. This seems to come from left field. This section needs to be re-written to clarify what is being done, and why.

Throughout the chapter the term buffering is used incorrectly. Buffering more specifically pH buffering is the resistance to change in pH. pH buffering can be high due to dissolution of aluminum minerals while contributing to acidification. A better term is the ability to neutralize acid.

4-53, paragraph- I strongly disagree with the statements on steady-state vs dynamic models. Agreed critical loads are a steady-state phenomenon, but ecosystems are not at steady state. A more balanced treatment should be given in the text. There are advantages and disadvantages associated with steady-state and dynamic models, which should be stated. Earlier in the text a dynamic model MAGIC is used for "critical load" calculations. (Although I might argue that these are not critical load calculations, rather target loads.) Application of steady-state models are problematic for ecosystems that are not at steady state. As many sensitive forest ecosystems are exhibiting soil exchangeable cation depletion, one might argue that it is not appropriate to use a steady-state model.

Specific Comments

4-1, line 15- Occult deposition is a poor term. I would just refer to cloud and fog deposition.

4-1, line 21- Change to: depleting soil exchangeable base cations…

4-2, line 2 and throughout the document- Change to: dissolved inorganic aluminum…

4-2, line 27- Need to define nitrogen saturation.

4-3, line 14- Change to: ecosystems and biological species…

4-4, line 1- Change to: available base cation pools of…

4-4, line 14- Change to: chronic or base flow chemistry….occasional acidic episodes, with..

4-4, line 41- Need to define base cation surplus.

4-5, line 21- The statement made concerning PnET is not entirely true. ANC can be calculated as base cations less strong acid anions in PnET-BGC, but PnET also simulates ANC by depicting the protonation of bicarbonate, organic anions and aluminum to represent measured ANC.

4-5, line 24- The statement made is incorrect and contradicts the statement made on line 13. Change to: Low ANC coincides with effects on…

4-13 on Figure 4.2-4 and several of the other figures in the chapter (e.g., 4.2-12, 13, 14) it is a challenge to read the labels and scale on the figures. The font size needs to be increased. The quality of these figures should be improved.

4-13, line 17 and throughout the chapter- Change to: trends in sulfate and nitrate concentrations and ANC…

4-14, line 3- Change to: were used...

4-14, line 6- Change to: because historical measurements are not available.

4-14, line 12- Change to: and low concern (Table 4.2-1).

4-16, line 14- Change to: 0 ueq/L (acidic), 20 ueq/L...

4-16, Table 4.2-1, first row Change to: Near complete loss of fish...

4-17, line 8- Change to have decreased (Figure 4.2-3)…

4-18, Figure 4.2-6- If these are mean values for all lakes this should be made clear.

4-18, line 11- Change to: base cation supply neutralizes the inputs…

4-18, line 17- It is not clear which monitored lakes is being referred to here; the LTM lakes?

4-21, line 9- Change to: less neutralizing ability…

4-21, line 15- Change to: the lake could neutralize and still…

4-29, line 2- Change to: have less ability to neutralize acid inputs than sites……

4-31, line 7- Change to: may degrade by 2050…

4-24, line 22- In addition to inputs of organic acids, some watersheds simply have low rates of base cation supply.

4-36, line 19- What is meant by accounting for effects of chloride?

4-37, paragraphs 2 and 3- This section is confusing. For example on line 21 it states maximum deposition load for sulfur is equal to the amount of sulfur the catchment can remove. On line 14 an assumption is that long-term sinks of sulfate in the catchment is negligible. This section needs to be re-written. I thought I understood the SSWC model until I read this section.

4-38, line 14- Change to: all possible combinations of…

4-38, line 17- Change to: each combination of depositions...

4-40, line 24- Change to: ANC = 50 ueq/L.

4-44, paragraph 1- You also may want to mention that regeneration of sugar maple is restricted under low calcium conditions. See Juice et al. 2006.

4-50, line 17- the ability of a system to neutralize acid…

4-50, line 18- Change to: successfully neutralize acidifying…

4-50, line 20- Change to: to neutralize acidifying…

4-50, line 23- Change to: bedrock with low ability to neutralize acid inputs..

4-55, line 16- This statement is not true. Watershed 6 has been cut and the forest biomass has been impacted by climatic disturbance events. See Aber et al. 2002.

Chapter 5

General comments

The nutrient enrichment chapter is well done. It is good to see a balanced description of the effects of fertilization in the beginning of the chapter. The Potomac and Neuse case studies illustrate the difficulty is quantifying the effects of atmospheric nitrate deposition in complex and diverse watersheds.

The chapter would benefit from a brief description of SPARROW, including the limitations of the model. Using a model like SPARROW will not allow for a determination of the time of recovery in response to decreases in nitrogen loading.

Specific Comments

5-9, line 14- Should the sentence say "at which the system is not nitrogen-limited"?

5-19, last paragraph- I don't understand incremental yield and delivered yield. Can these terms be clarified?

5-20, line 27- Define TNs.

Figures 5.2-7 and 5.2-11. There appears to be a mistake in the figure legend. Shouldn't the units be kg/ha-yr? Also more descriptive figure titles defining incremental nitrogen yield and delivered nitrogen yield would be helpful.

5-33, line11- Change to: levels by decreasing the oxidized…

Figure 5.2-16. Note that the concentration scale here is very small, not very meaningful differences in terms of measured concentrations.

5-45, line 18- Space between section 3.3). and Changes..

5-45, line 25- Typo: waters, reduced…

5-45, line 28- Typo: uptake (Figure 5.3-1).

5-48, line 27- Change to: from decreasing nitrogen…

5-58, line 18- Typo: analytical

Chapter 6

General comments

The material on the linkages between mercury and sulfur is generally a good overview. There are some inconsistencies in the text that need to be revised. For example on p -2, line 26 it is stated that inconsequential amounts of methyl mercury can be produced in the absence of sulfate. But on p 6-4, line 12 the text correctly indicates that methyl mercury can be produced by iron reducing bacteria. The first statement should be deleted. On p 6-3, line 5 the statement is made "Watersheds with conditions known to be conducive to mercury methylation have been identified in the northeastern U.S. and southeastern Canada.." but is followed by the statement " whereas watersheds with elevated methylmercury levels… are seen in most of the U.S. The first phrase should be deleted. It is inconsistent with the second part of the sentence and with figure 6.2-5.

Specific comments

6-1, line 24- Change to: stated that decreases in visibility…

6-3, line 17- Should be Driscoll et al. 2007

6-7, line 34- I believe now all 50 states have some sort on mercury consumption advisory.

6-20, line 16- There seems to be part of a sentence missing.

6-21, line 15- What other types of ecosystems were data available besides conifer and deciduous forests?

6-23, line 5- Standard deviation is given for N_2O and CH_4, can the variability for CO_2 uptake response factor be given.

6-23, line 26- Should this be section 6.4.4?

6-25, line 17- Change to: Because of decreases in ambient SO_2…

Chapter 7

General comments

Some of the overall suggestions that I made at the beginning of this report could be incorporated in this section.

7-23, line 22- The information presented in Table 7.1-3 is not quantitative and no information on Bc/Al is presented. This sentence needs to be changed.

Specific comments

7-2, Figure 7-1- I find the font difficult to read on the purple background in this figure.

7-5, Line 10- Change to (i.e., rain, snow), cloud and fog, and dry…

7-5, line 12- Change to: Both are essential. Nitrogen often limits the growth or productivity, and species diversity of ecosystems.

7-5, line 14- Change to: acidification and with excess nitrogen to nutrient enrichment.

7-5, line 16- Change to: localized loss or extinction…

7-7, line 22- Change to: nearly complete loss of…

7-8, paragraph 2- Again maximum depositional load. How is this related to critical load?

7-9, line 2- Change to: lake could neutralize inputs of acids and still…

7-10, line 1- Do you mean: 30 ueq/L in summer than in spring…

7-10, lines 2 and 3- Change drops to decrease.

7-10, line 3- Change to: Severe impacts can occur to fish…

7-10, line 5- Do you mean low or high pulses?

7-10, line 14- Change to: mobilization of dissolved inorganic Al…

7-10, line 25- typo on superscript.

7-10, line 26- Change to: concentrations of available Al, as measured by exchangeable base…

7-11, line 5- Need a period at the end of the table title.

7-11, line 12- Sverdrup and Warfvinge 1993 is not in the references.

7-14, line 21- Do you need the word magnesium?

7-17, line 19- red spruce

7-19, line 29- Is oconic a word? Iconic?

7-21, line 8- Change to: Tahoe drain through…

7-22, line 4- Are continuous monitoring units used for NOx?

7-22, line 16- Change to: data are…

7-23, line 7- Change to ANC and aquatic acidification.

7-23, line 8- Change to: between soil exchangeable Bc/Al ratio and terrestrial acidification.

7-23, line 21- Change to: between ANC and fish species…

7-23, line 22- The information presented in Table 7.1-3 is not quantitative and no information on Bc/Al is presented.

7-24, line 13- Change to: sustain terrestrial and aquatic food chains.

References

Aber, J.D., S.V. Ollinger, C.T. Driscoll, G.E. Likens, R.T. Holmes, R.J. Freuder and C.L. Goodale. 2002. Inorganic N losses from a forested ecosystem in response to physical, chemical, biotic and climatic perturbances. Ecosystems 5:648-658.

Juice, S.M., T.J. Fahey, T.G. Siccama, C.T. Driscoll, E.G. Denny, C. Eager, N.L. Cleavitt, R. Minocha, and A.D. Richardson. 2006. Response of sugar maple to calcium addition to northern hardwood forest. Ecology 87(5):1267-1280.

Dr. H. Christopher Frey

Comments on
"Risk and Exposure Assessment for Review of the Secondary National Ambient Air Quality Standards for Oxides of Nitrogen and Oxides of Sulfur, Second Draft"

GLOSSARY:

The definition of variability in the glossary is weak and misleading. The explanation of variability given on page 2-18 is much better. The glossary should be updated to better reflect accepted definitions of risk assessment terminology.

EXECUTIVE SUMMARY

Please define NO_y in the text box.

Figure ES-2: The term "atmospheric landscape" is confusing. One does not associate a landscape with the atmosphere. Use a more descriptive term.

Page ES-4: It is helpful to list what was analyzed. However, some explanation of what was not analyzed and why would also be helpful. For example, earlier it is indicated that sulfur is a nutrient, and a reader may wonder therefore why sulfur "enrichment" is not considered.

Figure ES-3. Remove the brown background color, because it is not useful.

Page ES-8. The "concept of ecosystem services" is mentioned but not defined. It should be explained to the reader.

Page ES-9 . The allusion to monetization in the first paragraph seems entirely inappropriate for this document. By law, the costs associated with a potential standard cannot be considered in setting the standard. Why ***should*** any of the endpoints be monetized in the context of this document? The issue of monetization related to benefits assessment, which may be relevant to a regulatory impact analysis or to information requested by OMB, but is not relevant to the process of developing NAAQS. Hence, either delete this material throughout the document, or provide a context that clearly differentiates that it is not related to standard setting and is mentioned for other reasons, and enumerate what those reasons are.

Page ES-10. Since CMAQ is a framework, it is non-specific to say that "CMAQ" was used to simulate concentrations. The reader would need to know more about what versions of components within CMAQ were used – i.e. what chemical mechanism, what advection algorithms, what treatment of planetary boundary layer, what "light" model, etc.

Figure ES-5. What layer of the atmosphere is represented here? Is it the lowest layer? Similar comment for Figure ES-6.

Figure ES-7. In lower right, it appears that there is a minus sign but some explanation should be given as to why "Total N – CMAQ dry + NADP Wet"

Page ES-14. For policy relevant background concentrations, please indicate how these were estimated: where they estimated based on monitored values at remote measurement stations, or are they based on model estimates?

Page ES-15. What was the finding regarding whether "area-based risk and exposure assessments are … suitable"?

Page ES-16. The level of detail on this page and a few others that follow seems to ramp up significantly. Who is your audience? If the ES is intended to be read by lay policy people, they will either stop reading here or eyes will glaze over. Try to write in shorter sentences and paragraphs. For example, the paragraph that starts on page ES-15 and goes onto page ES-16 is nearly a page long. The text box contains a run-on sentence and awkward and unclear definition of critical load. There is much repetition in the second paragraph on page ES-16. The phrase "ANC values" or "ANC limits" or similar is repeated three times.

Page ES-17. Top of page: "It was not possible to… a larger dataset" What larger dataset was desired? Larger in what way? This sentence is unclear.

"may aid" does this mean that the writer is not sure if the "connection" helps determine "adverse impacts"? Why so tentative?

The section non Terrestrial Acidification is likewise not likely to be read by a lay policy reader. Is the audience intended to be peer experts in terrestrial acidification? If so, they can probably read this. Doubtful that others can, however. May want to start out differently, explaining and introducing concepts step by step. i.e. soil acidification is bad, and explain why. Then explain that acidification of soil changes the concentrations of Ca++ and Al, and why. Then introduce that Ca++ is part of a "base cation" (explain what that is) that includes a few other ions. Then introduce the ratio of Bc to Al, and explain that it gets smaller or larger as acidification gets more severe. Etc.

Last paragraph on the page. Do the numbers 0.6, 1.2, and 10 have units? The way the sentence is written, it is not clear that these are Bc/Al ratios, since the sentence says that Bc/Al was calculated from the number given. Very unclear to the reader.

Page ES-18. What is a Al/Ca++ imbalance? In what way are Al and Ca++ supposed to be in "balance"? And why is the ratio now inverted? These kinds of inconsistencies are a good way to lose the reader.

Middle of page, the monetization seems entirely inappropriate. See earlier comment.

Page ES-20. Please clarify if "atmospheric deposition" includes only direct deposition or if indirect pathways are included – e.g., does deposition to land and then run-off into estuaries constitute "atmospheric deposition" to the estuary? It is a bit odd and confusing to the reader to say that more than 100% or greater reduction in atmospheric deposition was necessary. How can one have more than 100% reduction in deposition?

Bottom of page… e.g., 1.5 kg N/ha – should there be a time dimension to this number? i.e. over what time period did this amount of deposition take place?

Page ES-22: The term "mycorrhizal community" is mentioned before it is defined. This will confuse lay readers who are not experts in the topic area.

Table ES-3. Is the area really known to 6 significant figures?

Bottom of page ES-22. "The pressures exerted" seems to be a metaphor. In technical writing, avoid technically-based metaphors, as they can be confused for a literal interpretation, depending on the background of the reader. Since my background is mechanical engineering, I tend to think of pressure in terms of force per area. I don't think this is the intended meaning.

Page ES-24. It is not clear to the reader as to why the critically important role of N2O as a greenhouse gas is beyond the scope of this assessment. If they are beyond the scope of this "review," then why were they addressed qualitatively. It is contradictory to say they were beyond the scope but that they were included in the assessment, even if qualitatively. Why is an assessment of the effect of N2O on climate of necessity qualitative? Why can't it be quantitative? What are the key findings of the qualitative assessments of these endpoints?

There needs to be a section that addresses the key findings, conclusions, and implications associated with assessment of uncertainty and variability. What are the largest sources of uncertainty and variability? Given the uncertainty and variability, what findings can be made with a high degree of confidence? Which can't?

CHAPTER 7. My review focused on the section regarding uncertainty.

The section on uncertainty, starting on page 7-22, reads like the typical qualitative "laundry list" approach of acknowledging uncertainties but not characterizing them. This is, frankly, unacceptable.

The National Research Council (NRC, 1994) stated the need to describe uncertainty and to capture variability in risk estimates. Risk characterization became EPA policy in 1995, and the principles of transparency, clarity, consistency, and reasonableness are explicated in the 2000 Risk Characterization Handbook (EPA, 2000). Transparency, clarity, consistency, and reasonableness criteria require analysts to describe and explain the uncertainties, variability, and known data gaps in the risk analysis and imply that decision makes should explain how they affect resulting decision-making processes (USEPA, 2000, 1992, 1995).

On numerous occasions, the NRC has explicitly called for the use of probabilistic risk assessment (NRC, 2007a,b). NRC (1992) recommended that EPA should thoroughly discuss uncertainty and variability in the context of ecological risk assessment (NRC, 1993). NRC (1994), in a major review of risk assessment methodology, stated that "uncertainty analysis is the only way to combat the ‘false sense of certainty,’ which is caused by a refusal to acknowledge and [attempt to] quantify the uncertainty in risk predictions." NRC (2002) suggested that EPA's estimation of health benefits were not wholly credible because EPA failed to deal formally with uncertainties in its analyses.

EPA's Science Advisory Board has made recommendations similar to those of the NRC. Parkin and Morgan (2007) urged the Agency to characterize variability and uncertainty more fully and more systematically and to replace single-point uncertainty factors with a set of distributions using probabilistic methods (Parkin and Morgan, 2007). EPA has developed numerous internal handbooks on how to conduct quantitative analysis of uncertainties in various contexts (e.g., EPA, 1995; 1997; 1998; 2000; 2001) EPA (2009) provides a detailed overview of the current use of probabilistic risk analysis within EPA (including 16 detailed case study examples), an enumeration of the relevance of PRA to decision-making, common challenges faced by decision makers, an overview of PRA methodology, and recommendations regarding how PRA can support regulatory decision making. EPA's National Exposure Research Laboratory has recently explored methodological issues for dealing with uncertainty quantitatively when coupling models for air quality, exposure, and dose (Ozkaynak et al., 2008).

There are numerous texts on how to conduct analysis of uncertainty (e.g., Morgan and Henrion, 1990; Cullen and Frey, 1999; Vose, 2008). The World Health Organization has recently released guidance on qualitative and quantitative methods for uncertainty analysis in the context of exposure assessment (WHO, 2008). These guidelines have been used by EPA to support uncertainty assessments of exposure and health effects for criteria pollutants under the National Ambient Air Quality Standards (NAAQS). Hence, the framework is a general one. In particular, WHO proposed guiding principles that are adapted here:

- Uncertainty analysis should be an integral part of the assessment.
- The objective and level of detail of the uncertainty analysis should be based on a tiered approach and be consistent with the overall scope and purpose of the assessment.
- Sources of uncertainty and variability should be systematically identified
- The presence or absence of moderate to strong dependencies between inputs is to be discussed and appropriately accounted for.
- Data, expert judgment, or both should be used to inform the specification of uncertainties for scenarios, models, and inputs.
- Sensitivity analysis should be an integral component of the assessment.
- Uncertainty analyses should be fully and systematically documented in a transparent manner, including: quantitative aspects pertaining to data, methods, inputs, models, outputs; sensitivity analysis; qualitative aspects; and interpretation of results
- The results of the assessment including the uncertainty should be subject to an evaluation process that may include peer review, model comparison, quality assurance, or comparison to relevant data or independent observations.
- Where appropriate to an assessment objective, assessments should be iteratively refined over time to incorporate new data, information and methods in order to reduce uncertainty and improve the characterization of variability.
- Communication of assessment uncertainties to the stakeholders should reflect the different needs of the audiences in a transparent and understandable manner.

The decision context of risk assessment includes: (a) how to prioritize the activities of the assessment, and development of data for the assessment, in order to characterize and, where possible, reduce uncertainty; (b) how best to manage risk. Decision makers often want to know: who is at risk; the magnitude of risk; and tradeoffs between risk management alternatives. Examples of specific questions that decision-makers may ask include (Bloom et al., 1993; Krupnick et al., 2006):

- How representative is the estimate, (e.g., what is the variability around an estimate)?
- What are the major gaps in knowledge, and what are the major assumptions used in the assessment? How reasonable are the assumptions?
- Would additional data collection and research likely lead to a different decision? How long will it take to collect the information, how much would it cost, and would the resulting decision be significantly different?

Generally, from a scientific perspective, it is preferred to quantify uncertainties wherever possible. As WHO (2008) explains (p. 31):

> *Determination of an appropriate level of sophistication required from a particular uncertainty analysis depends on the intended purpose and scope of a given assessment. Most often tiered assessments are explicitly incorporated within regulatory and environmental risk management decision strategies. The level of detail in the quantification of assessment uncertainties, however, should match the degree of refinement in the underlying exposure or risk analysis. Where appropriate to an assessment objective, exposure assessments should be iteratively refined over time to incorporate new data, information and methods to reduce uncertainty and improve the characterization of variability. Lowest-tier analyses are often performed in screening-level regulatory and preliminary research applications. Intermediate tier analyses are often considered during regulatory evaluations when screening-level analysis either indicates a level of potential concern or is not suited for the case at hand. The highest tier analyses are often performed in response to regulatory compliance needs or for informing risk management decisions on suitable alternatives or trade-offs.*

Hence, the Tier 1 (Qualitative) approach is not a default. It should be a justified choice that is consistent with the purpose and scope of the assessment.

WHO specifies a structured approach to qualitative assessment of uncertainty that includes

1) qualitatively evaluate the level of uncertainty of each specified source;
2) define the major sources of uncertainty;
3) qualitatively evaluate the appraisal of the knowledge base of each major source;
4) determine the controversial sources of uncertainty;
5) qualitatively evaluate the subjectivity of choices of each controversial source; and
6) reiterate this methodology until the output satisfies stakeholders

Somewhat amazingly, the document seems to ignore an effort undertaken in the mid 1990s to create a quantitative framework for characterizing variability and uncertainty associated with acid deposition, which is the **Tracking and Analysis Framework (TAF)**. The details of this framework, and the associated software, are available at http://www.lumina.com/taf/. TAF represents a systematic effort sponsored by the National Acid Precipitation Assessment Program (NAPAP) to develop an "integrated assessment" model that represents emissions, transport and fate, deposition, and adverse effects to terrestrial and aquatic systems, and for which variability and uncertainty were quantified. The philosophy of TAF was to develop a reduced form model that was tractable and that could be used for repetitive analysis, such as "what-if" policy scenarios, sensitivity analysis, and assessment of the effect of variability and uncertainty on estimated end points. This approach is a practical one, and should serve as an illustrative example of how variability and uncertainty can be quantified in an assessment such as needed in the REA. EPA should review TAF and either use this framework, or update it and use it.

A laundry list of uncertainties, as given in Section 7.2 is not useful in interpreting the assessment results unless it is conducted in a systematic manner that puts priority on quantification and that leads to comparative assessments of which sources of uncertainty are important with respect to well defined endpoints. At a minimum, there should be summary tables that categorize sources of uncertainty with respect to a well defined endpoint. What are the implications of uncertainty for interpretation of the assessment results?

Why is there a benefits assessment component to Chapter 7? As noted in comments regarding the executive summary, costs cannot be considered when setting the NAAQS. What is the purpose of providing monetized assessment of benefits in the context of this REA?

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Dr. Paul J. Hanson

Final Comments on the Second Draft

Risk and Exposure Assessment (REA) for Review of the Secondary National Ambient Air Quality Standards for Oxides of Nitrogen and Oxides of Sulfur

Submitted 29 June 2009

General Comments:

I found the second draft REA to be improved over the first draft. The document is largely successful in defining air quality indicators and ecological indicators that might be used in the context of evaluating exposure metrics for both aquatic and terrestrial ecosystems in the context of acidification and nutrient enrichment for exposures to SO_x and both oxidizes and reduces forms of nitrogen.

In some cases data and justification are provided for the levels of the ecological indicators that might be considered in evaluating ecological responses, but for the most part levels, averaging times, and forms are not discussed.

While the document makes clear statements about the nature of exposures to US land surfaces and target case study ecosystems, it provides very little (if any) useful characterization of the welfare risks involved in allowing current pollutant levels to continue. Case studies for nitrogen deficient ecosystems were not included in the analysis.

The following specific comments and minor editorial suggestions are provided for discussion and consideration by EPA staff.

Front Matter – Key Terms

Page xxi: Add a definition for ASSETS

Page xxii: The definition listed for Determined Future Outlook doesn't stand on its own. You might also reference page numbers in the body of the text for all of the definitions related to ASSETS.

Page xxiii: Prior comments on the definition of ecological dose were not addressed. Why is this definition limited to microbes?

Page xxvi: The definition of a semi-arid region was not changed from the first draft. The rainfall amounts overlap with those for Arid Regions, which seems inappropriate.

Executive Summary:

Page ES-2 Line 12: Change "These effects include" to 'When fully developed acidification effects include…'

Page ES-2 Line 22: Change to " a well-documented phenomenon indicating…"

Page ES-3 Line 6: Change "the" to 'that'.

Table ES-7 under Terrestrial acidification: "Tree Health" is used as an undefined and unclear term. Use other words or phrases to describe what is really intended (e.g., changes in growth).

Figure ES-3 and elsewhere throughout the REA: Although some process was conducted to limit the number of case studies for inclusion within the REA that process is not well described. Why were these case studies chosen from an original longer list of possibilities? Should these be characterized as worst-case scenarios? Should the reader assume that these are the only areas of the US for concern with respect to acidification or nutrient enrichment?

Figure ES-4 and elsewhere throughout the REA: "Change in ecosystem structure and process" is used to describe an ecological benefit/Welfare effect. How do (or can) we distinguish changes occurring through natural processes from changes from the effects of acidification and nutrient enrichment? The document should include some discussion about ecological changes in the context of 'background temporal changes" vs. those driven by pollutant exposure.

Pages ES-11 to ES-14 are quite good, but I noted one issue. The term NOy is used in the caption for figure ES-5, but it hasn't been used much in the text. Is it intended to be a placeholder for total reactive forms of N?

Page ES-17 Line 18: In this paragraph sugar maple and red spruce are characterized as being the "most sensitive" to acidification with the implication that all other tree species are less sensitive. Is this really true? Perhaps other tree species simply haven't been evaluated in enough detail to appropriately characterize their sensitivity. Please reword the beginning of the paragraph to indicate that these species are being highlighted because sufficient data are available to evaluate their response to acidification.

Table ES-2: Does the concept of a policy relevant background Bc/Al ratio belong in this discussion?

Page ES-18 Lines 4 through 16: These paragraphs provide a description of valued characteristics of northeastern forests, but they **do not** provide an indication of the fraction of these welfare metrics that are at risk under acidification**.**

General comment: The previous statement is a recurring theme throughout the REA (especially in Chapters 4, 5, and 6). Ecological benefits or measurable welfare metrics are listed for key ecosystems or case study areas with the presumption that all are subject to loss or failure with acidification or nutrient enrichment. In most cases the text (and presumably the available data) do not provide sufficient information to fully characterize what fraction of a measurable ecological endpoint is likely to be subject to loss under pollutant exposure. I don't believe that it is appropriate for the reader to conclude that 100 percent of a given welfare metric is likely to be lost.

Page ES-21 Line 7: Don't use the term ecological health without an adequate definition.

Page ES-21 Line 12: Can you provide the deposition rate needed to drive mortality? Is it a higher level of deposition or might it alternatively be simply long term cumulative exposure to a lower deposition level?

Page ES-21 Lines 20 and 21: Please provide a range to clarify what is meant by a low C: N ratio.

Page ES-22 Lines 19 to 21: Surface area increases of root systems driven by mycorrhizae are most often associated with the morphological changes driven by ectomycorrhizae, but the authors are using AM as the example. Are they referring to fungal filament exploitation beyond the root systems of plants?

ES-23 Line 13: Are fishing and hunting really a big land use activity for the California Coastal Sage area?

Chapter 1:

Page 1-12 Line 19: Remove the word "not". It appears to generate a double negative that changes the sentence meaning.

Page 1-13: I suggest modifying the sentence by adding the underlined text as follows: "Both are essential elements for vegetation growth and development, and..

Page 1-14: Should "main source" be changed to 'main anthropogenic source'?

Page 1-17: These are all good policy relevant questions. Unfortunately, a number are not addressed within the REA.

Chapter 2:

Page 2-1: This section starts out with a great point – "response to pollutant exposures can vary greatly between ecosystems". Unfortunately, the REA doesn't fully address how to handle extrapolation of responses in case study areas to the balance of the US.

Page 2-1 Line 19: I would remove the word "and" in this line.

Figure 2.4-1: Please add more explanations to the figure caption. What do the arrow widths imply?

General Comment: How do we distinguish ecological effects of acidification and nutrient enrichment from other co-occurring and likely highly correlated pollutant exposures (e.g., ozone)? This issue should be fully vetted in the document.

Chapter 3:

Page 3-2 Line 18: Add the phrase 'other forms of reactive N' after NOx.

Figure 3.2-1: Increase the font size.

Figure 3.2-4: Increase the font size.

Figure 3.2-6: The figure caption uses NOy, but the related text on the previous page (3-11) exclusively discusses NOx. The authors should be consistent and define and use NOy appropriately or not at all.

Figure 3.2-10: The color scale in this figure was inappropriately changed from the scales used in the two prior figures. This isn't a big deal, but by changing the color scale a direct visual comparison isn't really possible.

Section 3.2.5: Should the concept of policy relevant background loadings or deposition levels be introduced and used?

Temporal trend data for atmospheric deposition of N and S forms through time is presented in Appendix 2. I would like to see this material presented visually in Chapter 3 to emphasize the history of control successes on SO_x and NO_x.

Chapter 4:

Section 4.3.1.2

This section is a good example of how an ecological indicator can be developed for the characterization of a response to acidification. Unfortunately, the other metrics described in the REA are not this clear.

Page 4-43: Is this a true general statement or should it apply only to sugar maple on susceptible sites?

Page 4-47 Lines 9 to 12: This sentence underscores a continuing theme. It is difficult to isolate and estimate the proportion of a given measure of welfare benefit attributable to acidification and nutrient enrichment. See also Page 4-48 lines 4 to 6. Given this reality, how do we proceed in the development of standards to protect welfare issues without a capacity to judge success or failure in the context of the target pollutants (or combined pollutants)?

Chapter 5:

Page 5-1 Lines 6 to 12: This sentence should also appear in the executive summary.

Page 5-6 line 6: Spell out and define NEEA.

Page 5-44 and Section 5.3.1.1: This section is very useful and helpful to the reader. I would add sentences to further describe why this discussion is limited to a couple of southern California case studies. Are other ecosystems in the US not impacted? Is there insufficient data to evaluate impacts in other ecosystems? For those that don't live in the region discussed, tell them why or how these results have meaning to broader pollutant exposures across the US.

Page 5-46 Lines 1 to 10: Staff should remind the reader that one size doesn't fit all at this point in the discussion. These data are quite good for lichens, but they may not have quantitative value for evaluating the response for other species or species in other regions of the US.

Section 5.3.1.3:

How is the reader supposed to interpret this information? What fraction of these services is at risk? Much of this information seems tangential.

Page 5-55 line 1: This bullet statement was discussing Eastern United States ecosystems. Do we have semi-arid lands in the eastern US?

Page 5-55 Lines 9 to 19: These statements seem to undermine the discussion. If we don't have the data, why are we having the extended discussion?

Figure 5.3-5: A solid connection of this graphic to N deposition isn't made. What fraction of fuel loadings leading to fire danger and frequency can be attributed to pollutant exposure as apposed to natural secondary succession?

Page 5-66 lines 21 and 22: This is a key point.

Page 5-66 line 24: Replace "tree health" with terms that describe what you really mean.

Chapter 6:

Page 6-13 Lines 28 and 29: This statement is written as though it would apply equally to all ecosystems. I'm not convinced that this would be true for all systems at similar time frames.

Page 6-23 Lines 20 to 22: This is an important statement. I'm glad to see it included here.

Chapter 7:

Page 7-10 Line 17: Change "most" to 'known to be'.

Page 7-14 line 27: Add the level of deposition needed to drive mortality.

Page 7-15 lines 17 to 26: Why the focus on these specific metrics for CSS and MCF? Are we to conclude that these are the metrics that are the best fit for all US ecosystems? Where is the discussion about the metrics needed for the development of a US national standard? Is a standard likely to be based on most sensitive systems in the west and then applied to all ecosystems?

Page 7-18 Lines 9 to 30: What is the point of this information? How is it be used? What fraction of each of welfare metric is at risk under terrestrial acidification? I would remove the bullets on this page and also on page 7-17.

Page 7-19 lines 1 to 3: Add some specifics.

Page 7-19 line 29: Should oconic be iconic?

Page 7-22 lines 28 to 30: Again. This is a very key conclusion. How do we use this conclusion in the extrapolation of case study data?

Dr. Rudolf B. Husar

SOx and NOx Risk and Exposure Assessment (REA) Second Draft

EPA is to be commended for the significant improvements of REA, Second Draft. EPA has responded effectively to many of the comments from CASAC. In particular, the inclusion of a section on the CMAQ model description and evaluation is a significant addition (Appendix 1-1), since so many of the key exposure estimates and conclusions depend on CMAQ.

Appendix 1: Description of CMAQ Applications and Model Performance Evaluation

Appendix 1-4, line 5.

'The purpose of these evaluations is to provide information on how well model predictions match the observed data on the regional basis'. Evidently, 'regional' in the context of the REA means the entire Eastern US and the entire Western US. Since the SOx-NOx pattern varies considerably within each model domain, this large scale aggregation makes it difficult to assess the regional model performance, ie. NE, SE, NW, etc.

The criteria for 'acceptability' of model performance based on comparability with photochemical model performances is somewhat dubious. It is my understanding that CMAQ ozone simulations have not improved significantly for the past decade. Hence, photochemical model performance is a rather poor metric for this REA. A more defendable criterion for the CMAQ model evaluation may be the performance on deposition estimates, particularly of nitrogen compounds. The CMAQ model performance for simulating nitrogen (NO_3+NH_4) deposition is modest. Nevertheless, the REA (Appendix 1-5, Line 2) states that 'The model performance results give us confidence that our applications provide a scientifically credible approach for the purposes of this assessment'. EPA's confidence in the nitrogen deposition pattern is not shared by this reviewer.

Chapter 6 Additional Effects

6.1 Visibility, Climate and Materials

According to the charge sheet, this chapter contains results from 'some qualitative analyses for the additional effects, including visibility, climate and materials…' Evidently, the section on visibility, climate, and materials was included at the recommendation of several committee members. However, this Section 6.1 does not reflect the result of any analyses, but merely points to PM Criteria Document as the source where aerosol effects on visibility, climate and materials are treated in detail.

Since EPA has not presented a concise summary of the welfare effects on visibility, climate and materials, I would recommend eliminating this section 6.1 and replacing it with a simple disclaimer such as the sentence in Page 6-2, Line 2.

Certainly, the current phrasing does not give justice to the role of sulfates and nitrates in visibility and climate. For example, (Page 6.1, Line 16) the REA states that impairment of visibility 'can result from atmospheric particulate matter (PM), which is composed in part of sulfate and nitrate..'. In reality, even sub-microgram concentration of ambient SO_4/NO_3 **does** result in impaired visibility.

Also, the statement that 'theoretical and empirical findings suggest that sulfates often dominate the fine particle mass and hence the impairment of visibility' is a dubious formulation of the role of sulfates. The findings on the optical effects of sulfates is not just suggestive, but it is based on firm, direct measurements of both sulfate concentration and light scattering.

6.2 Sulfur and Mercury Methylation.
This section is relevant to the REA. It also properly illustrates the interaction of sulfates with the chemistry of other compounds.

6.3 Nitrous Oxide
The discussion of nitrous oxide is relevant to the climate effects of NO_x. This section appropriately describes the role of nitrous oxide. Unfortunately, the climate effects of sulfate/nitrate aerosols has fallen through the cracks. At a minimum, the recognition of those effects as part of the welfare effects of SO_x/NO_x should be stated along with the pointer to the Climate Section of the PM Criteria Document.

Dr. Dale W. Johnson

Review of "The Risk and Exposure Assessment for Review of the Secondary National Ambient Air Quality Standards for Oxides of Nitrogen and Oxides of Sulfur, Second Draft"

Amended 28 July 2009

Case Study Questions to the panel:

1. *Are the uncertainties appropriately characterized across the case studies? Is there adequate information to allow us to weigh the relative strengths of each case study to inform the standard setting procedure?*
2. *In using the Risk and Exposure Assessment to inform the policy assessment, we plan to focus on aquatic acidification as the basis for an alternative multi-pollutant secondary standard as this is the area where we have the most confidence in our ability to characterize adverse effects. Does the panel agree with this approach?*

My answers to these two questions are as follows, with more detailed reviews below:

1. No, the uncertainties are not appropriately characterized and there it not, in my opinion, adequate information to inform standard setting procedures. For the aquatic case studies, there is confusion about how anions and cations interact and considerable uncertainty as to how reduced (or increased) mineral acid anions like SO4 and NO3 affect base cation concentrations directly and immediately through the necessity for charge balance as opposed to much longer term change changes in soils. The document appears to implicitly assume the latter, but without soil data to verify it, we cannot know whether soils have in fact changed at all. For the terrestrial case studies, I believe that there are significant and erroneous assumptions in the simple model used that at least should be acknowledged and discussed before any conclusions are drawn. Also, the case for red spruce decline due to acidification is not as clear as the document would lead us to believe.
2. I think this approach is OK as a <u>focus</u>, but I do not believe that it should occur to the exclusion of all other effects. I assume that this will not be the case.

There are three chapters that deal with case studies: 3, 4, and 5. These case studies all appear to focus on sites either with high levels of deposition of very high sensitivity to increased deposition. This is logical, given that the focus of this report is on potentially negative effects, but care must be taken not to extrapolate the results from these case studies to regional or national scales without first accounting for the many other sites that are not sensitive.

Chapter 3 deals with deposition rates and characteristics of the case study areas, and I really have no comment or issue with anything there.

Aquatic Acidification Case Studies

Chapter 4 deals acidification, 4.2 deals with aquatic sensitivity and 4.2.3 focuses on the Adirondack and Shenandoah sites for case studies. Here, I would comment on the section on page 4-18 that it should include a more detailed discussion of the two possible reasons for the observed decline in streamwater base cation concentrations: 1) the reduction in the concentrations of mineral acid anions (mainly sulfate), which necessitate a reduction in cations, including base cations, in order to maintain charge balance, and 2) soil acidification. This relates to my earlier review comments as to the critical importance of considering capacity effects (change in the soil, which take a long time and are not easily reversed) and intensity (change in solution, which can take place almost instantly and are very easily reversed). The statement on lines 1-13, page 4-18 implicitly assumes that the changes are of the capacity nature, and ignore the intensity component, which in fact must play a role. Indeed, without soil data, it is impossible to know if capacity changes played any role at all or whether “”base cations buffer the inputs of NO3- and SO42-, which will likely limit future recovery of ANC concentrations.” This is a somewhat confused (cations do not buffer anions) and very incomplete statement, because if the base cation decline is due to changes in SO4 and NO3 only, complete ANC recovery can be expected if the levels of those anions are brought back to pre-industrial levels somehow.

I have significant problems with the premises upon which the equations and associated text on page 4-37 are based. First of all, the assumption that the preindustrial rates of base cation leaching are sustainable is completely without foundation; were this the case, we would never find acidized soils in nature (without pollution), and we certainly do find them. The natural genesis of soils with more rainfall than evapotranspiration is to acidify; the question here is to what extent this process has been sped up by pollutant inputs. Secondly, I think that the assumption that nutrient cycling effects by plants can be ignored is deeply flawed, as the literature is full of examples where tree uptake of base cations well exceeds the removal of base cations by leaching. Furthermore, it appears that N cycling is NOT ignored, so this is only a partially imposed and inconsistent assumption.

Specific Comments:

p. 4-25: The legend on Figure 4.2-14 is way too small
p. 4-36, lines 10-12: This is true only in acid soils. In basic soils, inputs of SO4 and NO3 will have little or no effect on the “acid balance of headwater lakes”. Anions affect total cation concentration by simple charge balance requirements, but they do not prescribe the type of cation.

Terrestrial Acidification Case Studies

Before specifically going into my assignment, I must comment on Section 4.3.1.1, Ecological Indicators. Here for the first time I see Bc/Al ratios, where Bc includes Ca, Mg, and K. This is only logical if the units are in moles or micromoles of charge (μmol_c), yet no units are given. What are the units? This appears to be based on a report by Sverdrup and Warfvinge published in 1993, but I was unable to get the reference and do not know what it is based on. The more commonly used indicator is that Ca/Al molar ratio of Cronan and Grigal (Cronan**,** C.S. and D.F. Grigal**.** 1995. Use of Ca/**Al** ratios as indicators of stress in forest ecosystems. Journal of Environmental Quality 24: 209-226.

Also, on page 4-44, it is clearly stated that acidification negatively affects red spruce – I admit to not being up to speed on the latest developments, but I do recall that Art Johnson found that climate change, not acid rain, was responsible for the red spruce decline in New England. I think this treatment is a bit one sided and the situation is not that clear. (Johnson, A.H., E.R. Cook, and T.G. Siccama. 1988. Climate and red spruce growth and decline in the northern Appalachians. Proc. NatI. Acad. Sci. USA, Vol. 85, pp. 5369-5373.)

The actual case studies for HBEF, which assumes that net forest increment is zero (I doubt that very much, even if the site has not been recently harvested and would ask for some documentation of that) and the KEF sites, and builds the analysis on the SMB model which has numerous assumptions and the unitless Bc/Al ratio to conclude that "These results suggest that the health of red spruce at HBEF and sugar maple at KEF may have been compromised by the acidifying nitrogen and sulfur deposition received in 2002." I realize that all models are imperfect and yet there may be good reasons to run them anyway, but this seem grossly overstated and I would add many many caveats to this section.

Nutrient Enrichment Questions for the Panel:

1. *Section 5.2 and Appendix 6 describe the analyses used to evaluate the effect of aquatic nutrient enrichment. The analysis uses the SPARROW model on one stream reach (Potomac River and Neuse River) to determine the impact of atmospheric total nitrogen deposition on the eutrophication index for the estuary. Does the Panel think that the model is adequately described and appropriately applied?*
2. *Section 5.3 and Appendix 7 describe the analyses used to evaluate the effect of terrestrial nutrient enrichment. This qualitative analysis describes the impacts due to nitrogen deposition on the Coastal Sage Scrub community of California and mixed conifer forests in the San Bernardino and Sierra Nevada Mountains and larger areas where possible. In addition, the effects on nitrogen deposition in the Rocky Mountain National Park supplemental case study location are summarized. How would the Panel apply threshold values presented in this case study to allow for broader geographic application that accounts for regional variability? Have the associated uncertainties been adequately characterized?*

My answers to these two questions are as follows, with more detailed reviews below:

1. I cannot intelligently answer this question. I am not familiar with the SPARROW model and would not be confident in commenting on it without considerably more information as to its structure and premises – more than could or should be included in a document such as this. I will pass on this one to other panelists who probably have more knowledge on the matter than I do.
2. I would not apply the threshold values presented in this case study to a broader geographic region because these case studies, while appropriate for negative effects of pollutant inputs either at high levels or on sensitive sites, do not in any way address the larger majority of ecosystems which are either resistant to negative effects or in fact might benefit from the additional nitrogen inputs. This is in essence a philosophical issue: should standards be set on the basis of the most sensitive or highly impacted systems or

should they be based on regional effects? Since the question asked about regional application, I am assuming the latter and thus my answer is that these case studies are in no way regionally applicable. The uncertainties with this are considerable and not well characterized.

First of all, this section is really mislabeled: it really focuses on nutrient excess, not enrichment and I suggest Nutrient Excess as the title. Nutrient enrichment to terrestrial people often implies something good, and that certainly is not the focus of this section.

Aquatic Nutrient Enrichment

I am not familiar enough with either the SPARROW model or the sites to intelligently comment on that section.

Terrestrial Nutrient Enrichment

The case studies for nutrient excess are good ones, with excellent research programs documenting negative effects of high levels of N deposition. As in my previous reviews, however, I must again go to the mantra of taking a balanced approach to this issue and if this section is still to be entitled Nutrient Enrichment, some mention of the possible benefit to commercial forests in the Pacific Northwest and Southeast should be mentioned. I fully recognize by now how loathe the authors are to do this, but I will continue to make this comment as long as I am on this panel as I think it is important and the omission of it will be greatly regretted later. (for example, see Chappell, H.N., D. W. Cole, S. P. Gessel and R. B. Walker. 1991. Forest fertilization research and practice in the Pacific Northwest. Nutrient Cycling in Agroecosystems. 27: 1385-1314; see also this link for a lay article on fertilization in southeastern pine forests: http://www.ipni.net/ppiweb/bcrops.nsf/$webindex/2476B56D4FDD9EB0852571B1006A6F2E/$file/06-3p12.pdf)

Additional Effects Questions for the Panel

1. In this chapter, we have presented results from some qualitative analyses for additional effects including visibility, climate and materials, the interactions between sulfur and methylmercury production, nitrous oxide effects on climate, nitrogen addition effects on primary productivity and biogenic greenhouse gas fluxes, and phytotoxic effects on plants. Are these effects sufficiently addressed in light of the focus of this review on the other targeted effects in terms of the available data to analyze them?

My answers to this questions are as follows, with more detailed reviews below:

1. The segments on visibility, climate and materials, the interactions between sulfur and methylmercury production, nitrous oxide effects on climate seem to be adequately addressed, but, for the reasons given in detail below, the segment on nitrogen effects on primary productivity still has some serious problems and issues, as detailed below.

I have serious problems with the assessment of nitrogen effects on primary productivity in this section. First of all, the authors appear to go through significant intellectual gymnastics in order to either ignore or disprove the concept that greater primary production leads to more C sequestration. There may be cases where primary production does not lead to increased C sequestration, and the best example of this is the forest floor, which decreases in mass as mean annual temperature increases despite the increases in primary productivity. However, given that organic C for sequestration is produced during primary production, to cling to the notion that the two are not related defies logic.

Furthermore, the statements regarding nitrogen in this section are largely untrue and often given without citation, and fly in the face of published literature. Examples of this is are on page 6-13, lines 15-17 which states that growth increases due to N inputs are offset by increases in soil respiration and on page 6-14, lines 27-29 where it states that “increased leaf N concentration under conditions of elevated nitrogen deposition may result in higher carbon loss by increasing both autotrophic and heterotrophic respiration”. This is simply not true: most studies show that N fertilization causes **decreases** in soil respiration (see for example Tyree, Michael C.; Seiler, John R.; Fox, Thomas R. The Effects of Fertilization on Soil Respiration in 2-Year-Old *Pinus taeda* L. Clones Forest Science, Volume 54, Number 1, February 2008 , pp. 21-30; see also Olsson, P., S. Linder, R. Giesler, and P. Högberg. 2005. Fertilization of boreal forest reduces both autotrophic and heterotrophic soil respiration. Global Change Biology 11 1745– 1753; and Google “Fertilization effects on soil respiration” for many other references**)**

On page 6-14, lines 23-31, the authors state that higher nitrogen concentration in organic matter stimulates decomposition; while this is sometimes true in the early stages of decomposition, the literature is clear on the long-term effects: greater N concentration **increases** the long-term storage of stable organic matter.

See:

B. Berg and C. Mcclaugherty (2003). Plant Litter–Decomposition, Humus Formation, Carbon Sequestration. Springer Verlag, 286 pp., 76 figs.

Sarah E. Hobbie (2008) NITROGEN EFFECTS ON DECOMPOSITION: A FIVE-YEAR EXPERIMENT IN EIGHT TEMPERATE SITES. Ecology: Vol. 89, No. 9, pp. 2633-2644

Berg, B., Ekbohm, G., Johansson, M.-B., McClaugherty, C., Rutigliano, F., and Virzo De Santo, A. 1996. Maximum decomposition limits of forest litter types: a synthesis. Can. J. Bot. **74**: 659–672.

Berg, B., McClaugherty, C., Virzo De Santo, A., and Johnson, D. 2001. Humus buildup in boreal forests: effects of litterfall and its N concentration. Can. J. For. Res. **31**: 988–998.

For the reasons given above, I believe this section to be seriously biased and flawed. I believe that the authors need to better research the literature and present a more balance and accurate assessment of the effects of N on ecosystem C sequestration.

Amendments, 28 July 2009

<u>Executive Summary</u>

p. ES-15, lines 5-7 (only odd line numbers are shown?): The first sentence on this page is incomplete and misleading – if the mere deposition of Sox, NOx and NHx leads to exposure to acidification, then is the goal to have no deposition at all? If so, many ecosystems would have no N or S in them at all, as atmospheric deposition is the only source (there are no primary soil minerals containing N and few containing S). You should add the word “excessive” in front of “Deposition”.

p. ES-15, lines 7-12: The next sentence may well be true, but I am unsure if the ISA documented the fact that N and S deposition has increased soil N and S contents. And if so, there is nothing really harmful about this (as I think the authors would imply). All those follow on statements about leaching etc. refer only to specific cases where N and S deposition are excessive; they are NOT the general rule.

p. ES-17, lines10-11: Again, it makes no sense to use Bc/Al ratio to represent Ca/Al, when in fact Bc includes Ca.

p. ES-18, line 4: Now we are using Al/Ca? Be consistent.

p. ES-18, lines 19-20: The term “nutrient enrichment” does not imply nutrient imbalance at all, it implies exactly the opposite and will certainly confuse people. I suggest that you use the term “nutrient excess”

p. ES-19, lines 19-20: I thought that the coastal eutrophication problem was driven primarily by animal waste?

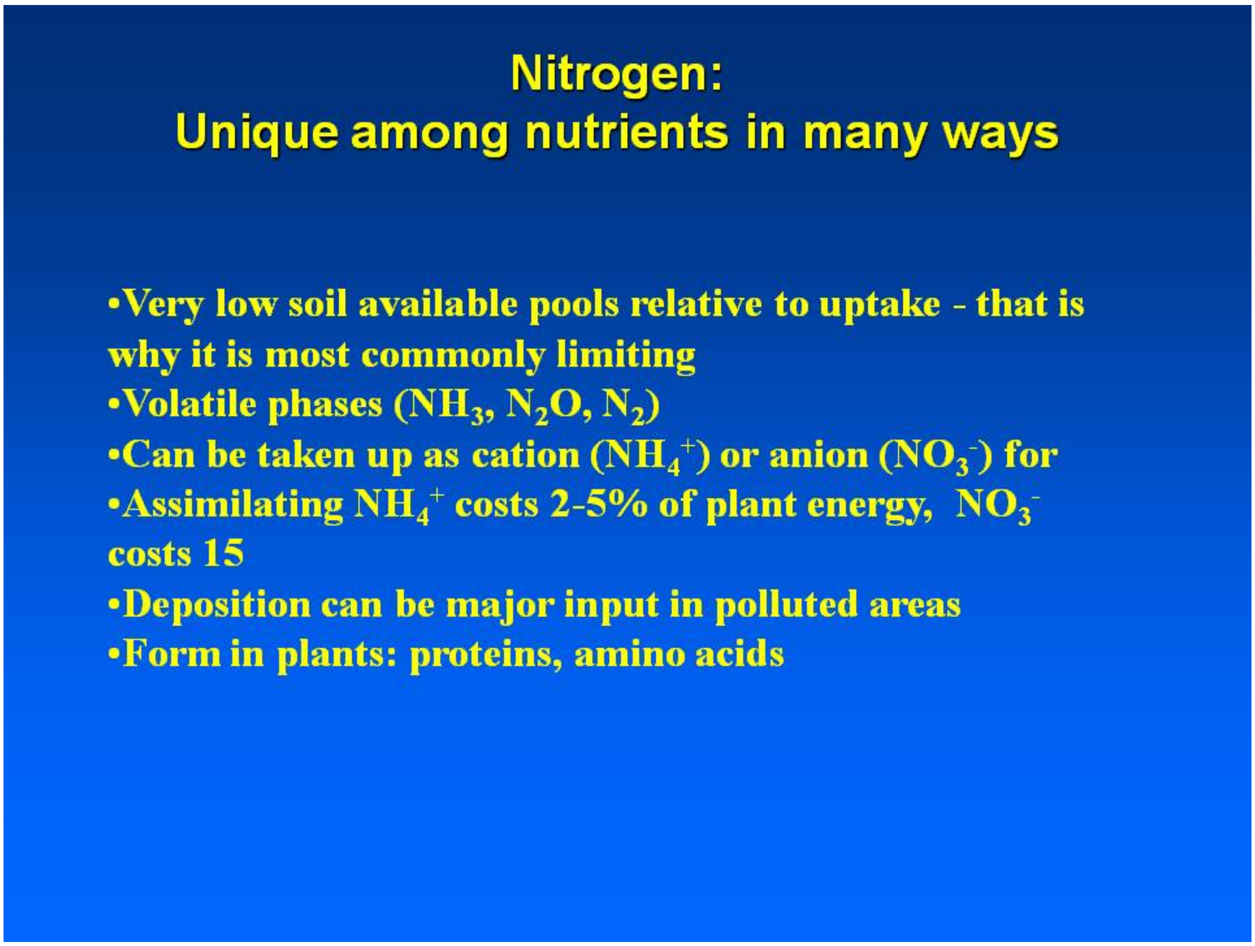
p. ES-21, lines 4-6: Now this is a good way to state the issue! It clearly states that excess nitrogen causes these problems and it certainly does! The rest of the ES and the entire document should reflect this – it is not a matter of nitrogen being “bad” , it is a matter of how much nitrogen is being applied.

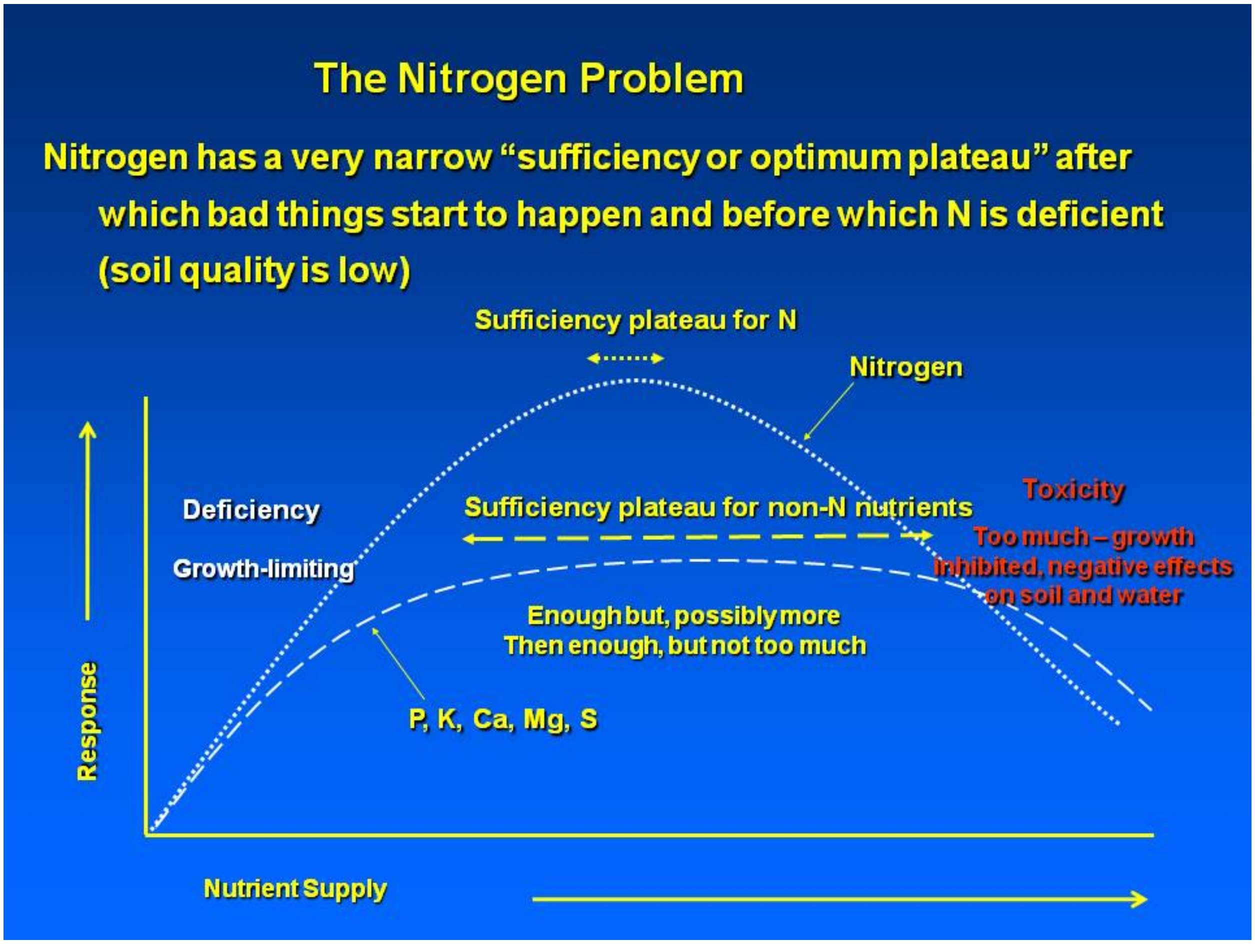
p. ES-21, line 19: Again, good wording: overenrichment is exactly the issue!

p. ES-24, lines 9-11: I do not share this confidence aside from the case study areas where N deposition is very high and ecosystems are sensitive. The Pregitzer paper that I asked to be reviewed in earlier comments shows the opposite effect. Again, I seek balance here. Not all areas are sensitive to current levels of N deposition – in fact, very few are. This summary and the document do not acknowledge this fact and it will be regretted later.

General Comments

At the suggestion of Ellis Cowling and Charlie Driscoll, I am inserting the following background material taken from my class presentation (NRES 497/697, Forest and Range Soils, Department of Natural Resources and Environmental Science, University of Nevada, Reno).




The Nitrogen Problem
Nitrogen has a very narrow "sufficiency or optimum plateau" after which bad things start to happen and before which N is deficient (soil quality is low)
Sufficiency plateau for N
Nitrogen
Deficiency
Growth-limiting
Sufficiency plateau for non-N nutrients
Toxicity
Too much – growth inhibited, negative effects on soil and water
Enough but, possibly more
Then enough, but not too much
P, K, Ca, Mg, S
Response
Nutrient Supply

The Nitrogen Problem

Nitrogen is the most frequently limiting nutrient and a high quality soil must have adequate N.

However, it is very difficult to manage nitrogen at an optimal level for plant growth while at the same time maintaining water quality and not causing negative effects on other soil nutrients (and causing deteriorating soil quality)

This is because soil mineral N pools (NH_4^+ + NO_3^-) will not remain elevated in soils for prolonged periods

- Although NH_4^+ is strongly absorbed by soils, high levels of NH_4^+ will stimulate nitrification, and NO_3^- will leach
- High levels of NO_3^- will leach (NO_3^- is poorly adsorbed to soils)

Dr. Donna Kenski

Revised comments, NOx/SOx Secondary REA (2nd Draft)

General Comments: Overall, the 2nd draft REA provides, through its case studies, a comprehensive summary of acidification and nutrient enrichment effects of sulfur and nitrogen deposition in sensitive ecosystems. I am generally satisfied that the data presented here sufficiently demonstrate the environmental damage done at current atmospheric concentrations of NOx, SOx and NHx and that it constitutes enough evidence on which to base a standard. What's still lacking is a clear discussion of the specific route to formulating a standard. The Executive Summary lays out a very general outline, which is repeated in various fashions in Chapters 1, 2, and 7, but there is still no concrete, detailed example of how a standard might be structured, despite having all the necessary information collected here. The committee raised a number of questions related to this in the last review, and it would have been good to see those addressed here, rather than waiting for a separate policy assessment. For example, how will varying degrees of geographic susceptibility and variability be incorporated (i.e., ANC might be the most logical indicator for the eastern US, but would it protect CSS and MCF communities?) Is EPA contemplating a standard based on just one of the ecological indicators (ANC seems like the logical choice) or will it be combined with other indicators? What kind of overlap might there be if multiple indicators are selected and how do we determine which is controlling? Is there any accommodation for uncertainty in the estimation of concentrations, deposition, ecological effects, and indicators? Because linking all of these modeled values will incorporate so many different estimates of variability and uncertainty, it is very difficult to imagine their cumulative effect on a standard. Given that EPA is proposing a brand new approach to an ambient air quality standard, the sooner these issues are dealt with and reviewed the better.

I like the stylized graphic of Fig. ES-2 that is used on the introduction pages to each chapter. The use of boldface for figure and table references in the text was a big help to readers hunting for those. Also I thought the use of the call-out boxes was very effective. Throughout the document, but especially in Chapter 3, the figures were well done – no extraneous information, thoughtful and consistent use of color. Very nice.

One point that needs to be made more strongly throughout the document, whenever the various ecosystem services are being described, quantified, or otherwise valued, is that they are only a small subset of the sum total of services we derive from these resources.

Exec. Summary: The last statement of the conclusions to the ES was disappointingly weak ("…effects due to aquatic and terrestrial acidification *may be* the most useful…"). The REA was quite convincing that the aquatic nitrogen enrichment effect was not going to be a suitable basis for a standard, and also that aquatic and terrestrial acidification were the effects we have the best science for and the most developed quantitative relationships on which to base a standard. The ES could be a bit more forthright about its findings.

Chapter 1: I like the list of policy relevant questions at the end of Chapter 1, but I expected to see answers to the questions when we got to the final synthesis and integration in Chapter 7. A conclusions section framed in such a way would be ultimately more useful than the current structure of Chapter 7.

Chapter 2: Good overview.

Chapter 3 (and Appendix 1): Great graphics, except for the unnecessary 3d pie charts. Conclusion 6 on p. 3-79 that the season (in the East) with the most TN deposition corresponds to the season with the most S deposition is difficult to confirm because the N and S data aren't shown together. Could a plot or two be added to demonstrate that this is indeed the case? The CMAQ comparison with measurements was well done and did provide some reassurance that CMAQ was adequate for this assessment, although the model performance assessment in Appendix 1 wasn't exactly what I was expecting. Making the assessments on an annual or monthly domain-wide basis isn't the typical way of evaluating CMAQ, and looking at those long term averages tends to make performance look much better than on a shorter term basis. If the logic was to look at long term averages because the case studies are only looking at long term averages, the text should be clear about it, although I'm not sure that's a sufficient reason. EPA has plenty of model performance guidelines, so it's surprising that the validation results presented here did not follow those guidelines. Also, I'm not convinced that because these results fell within the range of other studies, that constitutes acceptable performance (as discussed much too briefly on pp. 1-4 and 1-5). The performance is not particularly good for many of the components and it deserves a more thoughtful discussion of what is really acceptable for this particular application. That said, I think this is a much better approach than the previous draft's RSM approach.

Chapter 4: This chapter's finding that aquatic acidification in both case study areas was driven primarily by sulfate was not brought out in any of the conclusions or summary statements, although it seems like it could potentially be significant information in the standard setting process. I was confused by the contradiction between these statements (on pp. 4-18 and 4-25) and Figure 4.2-23, which implies that the two Adirondack lakes shown are sensitive to both sulfate and nitrate. These contradictory results would lead policy makers to very different conclusions, depending on which one is given the most weight. The text must do a better job explaining the reasons for the different conclusions (different models used, with very different assumptions), or otherwise reconcile these findings. The section (4.2) that looked at recovery potential should have examined at least one or two scenarios with reduced emissions, since maintaining emissions at current levels for the next 10 or 40 years is not realistic and expected emissions reductions might well make a significant difference in the estimated recovery times. It could have been tied in with the emissions reductions analysis in Chapter 3. Also, the chapter could use a brief discussion of the averaging time for ANC. It uses mostly annual average values, although frequent mention of episodic pulses of acidity are made. Do seasonal patterns in deposition have any impact on ANC? This isn't my area of expertise, coming from the ambient air world of hourly measurements, but it was a little startling to see that much of the lake data is based on one measurement a year. So a brief explanation of ANC behavior over time might be helpful. With respect to uncertainty, the specific discussions about uncertainty in the various models employed was generally adequate (note comments above about CMAQ though). The discussion on uncertainty in the aquatic nutrient enrichment was especially well done (Sec. 5.2.8). However, a broader discussion that pulls together uncertainty from MAGIC, along with uncertainty from CMAQ, and uncertainty in the effects of ANC on sensitive biota, etc., needs to take place somewhere. Chapter 7 takes a broader look but still doesn't discuss the cumulative, quantitative impact of these various sources of uncertainty on a standard. Perhaps this is more appropriately left for the policy analysis, but it needs to take place at some point.

The 2nd charge question for this chapter asks if we agree with focusing on aquatic acidification for a basis of the standard. I do agree; the quantity and quality of data available for aquatic acidification really make it the only logical choice, relative to the other effects examined in the REA. The nutrient enrichment studies were just too qualitative to think about basing a standard on them. However, it is important for EPA to continue to consider terrestrial acidification as well as the nutrient enrichment cases, and find a way to truly integrate multiple indicators into a standard. Multiple indicators will expand the geographic relevance and scope of the standard and afford protection to a maximum number of sensitive ecosystems.

Specific comments (page, line number)

ES-10, 10-11: MM is not an SI unit, nor is it a standard abbreviation in the air quality community. Scientific notation would be better. Or at least include it in the table of abbreviations.

ES-10, 18: fix subject-verb agreement

ES-15, 11: missing a closing parentheses for the phrase that starts "(especially…"

ES-16, 27, 36, and others throughout document: should be "sensitive *to* or at risk *from* acidifying deposition" . The phrase "at risk to acidifying deposition" is jarring.

ES-16, 31; ANC level ~~of~~ above

ES-18, 1: Bc/Al level ~~of~~ above

1-12, 17-19 : Is this sentence correct? Or do you mean there is little new evidence that S and N oxides are high enough to be phytotoxic (delete *not*?)

1-17, bullet 2: this bullet is awkwardly phrased – reword

2-2, 5: It is not clear what the role of these supplemental study areas is. Little Rock Lake, for example, is not summarized in Table 2.1-1 or Fig. 2.1-1, and it's not even mentioned again except a passing reference to it in Chapter 6. It hardly seems to merit mention here. Rocky Mountain alpine lakes are discussed in Chap 5.

2-7, 17: public

3-2, 9 and 18: the subscript x should be NHx

3-4, 1-4: MM is not an SI unit, nor is it a standard abbreviation in the air quality community. Scientific notation would be better. Or at least include it in the table of abbreviations.

3-11, 27: Figure 3.2-6 says it shows NOy, not NOx – which is correct?

3-28: Table 3.3-1 says the Neuse River has 14 kg N/ha/yr in 2002, but the figure 3.3-1a says it has 15; shouldn't these be the same?

3-30, 1: contains should be contain

3-82, 5 and 8: seems to be missing NH before the orphaned subscripts

3-90, 14: …are based *on* the application of

3-91,24: …the formation *of* sulfate and nitrate…

3-93, 5: similarities *and* differences

4-6, 11-14: edit this final sentence

4-11, 15: delete sulfate

4-23, Fig. 4.2-13 misspells exceedances in each of the 4 titles

4.34, 10-13: This first sentence is too long and convoluted.

4-37, 10: Shouldn't Q be in m3/yr?

4-39, 6: SSWC is not defined in the text or list of abbreviations – I had to dig in the Appendix for it.

4-40, 23: levels -> level

4-41, 3-12: This explanation of the MAGIC calibration procedure is pretty fuzzy. At least refer the reader to Attachment A of Appendix 4 for additional information.

4-45, 6-8: Delete these lines

5-16, Fig. 5.2-3: this figure was pretty confusing – too much information is presented, and it's hard to read besides.

5-47, Fig 5.3-2: the 7th bullet on Minnesota grasslands doesn't list any ecological effect. Farther down the list, Bytnerowicz is misspelled

7-12, 4: res -> red

7-16, Fig. 7.1-3: the 7th bullet on Minnesota grasslands doesn't list any ecological effect. Farther down the list, Bytnerowicz is misspelled

7-17, 19: res -> red

7-19, 29: oconic -> iconic

7-21, 12-16: fix run on sentence

Dr. Naresh Kumar

The second draft of the Risk and Exposure Assessment (REA) document is much improved from the first draft. However, there are still some areas that need improvement and further analyses that need to be performed. My major comments, with a main focus on the air quality analysis, are:

1. Figure 1.4-1 (reproduced below) appears as the key figure in the REA document. As noted, the Atmospheric Deposition Transformation Function depicted in Box 3 "quantifies the relationship between atmospheric concentrations and deposition of NOx and SOx".

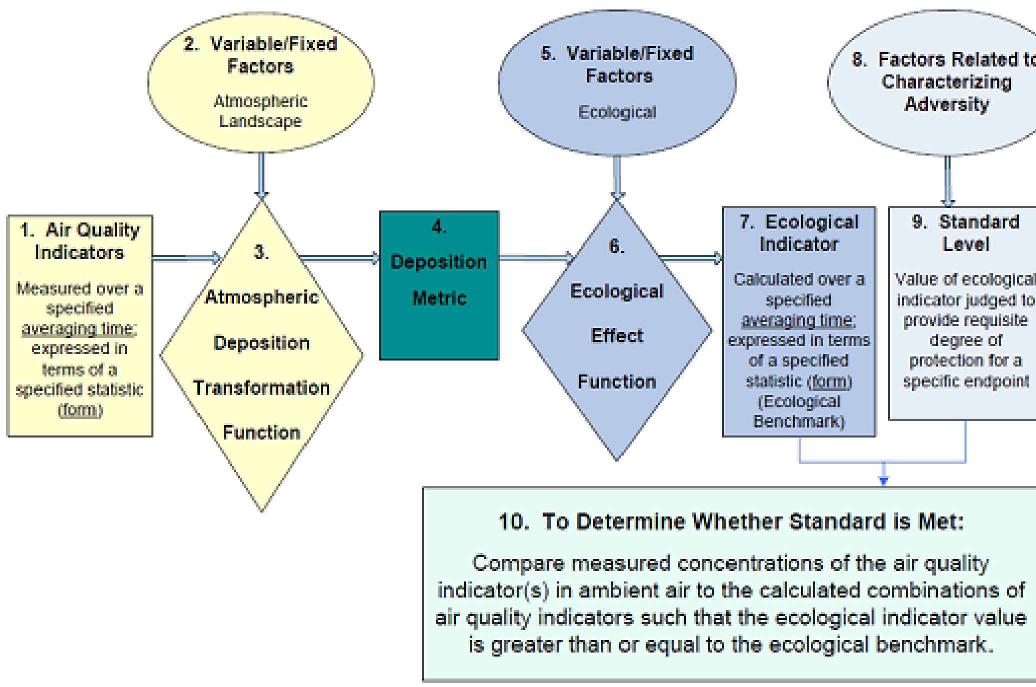


Figure 1.4-1. Possible structure of a secondary NAAQS for NO_x and SO_x based on an ecological indicator.

It appears that the CMAQ model used in the analysis is the "Atmospheric Transformation Deposition Function." However, the CMAQ model transforms emissions, not concentrations. Moreover, it predicts both concentrations and deposition fluxes. As a result, there must be some other form for the atmospheric function depicted in Box 3 that EPA has not shown in the document. The relationship between concentrations and deposition fluxes of NOx and SOx is expected to be proportional, i.e. changes in concentrations would be reflected as proportional changes in deposition fluxes. However, it is likely that the relationship will vary seasonally and spatially depending on regional chemical regime, precipitation and other meteorological variables, as well as land use and terrain features. Because of limited measurements of NOx, it would be difficult to show the relationships between observed concentrations and deposition, but (at a minimum) the CMAQ model results could have been used instead (as they are being used for dry deposition).

It is essential that EPA should show the relationships between concentrations of SO_2 and NOx and the corresponding deposition measures for different regions of the country and for different seasons. These should include:

a. The relationship between measured SO_2 and measured wet sulfur deposition
b. The relationship between modeled SO_2 and total sulfur deposition (measured wet deposition and modeled dry deposition)
c. The relationship between measured NO_2 and measured wet nitrate deposition
d. The relationship between modeled NOx and total nitrate deposition (measured wet deposition and modeled dry deposition).

The EPA should also explore the impacts of combined reductions in SOx and NOx on total N deposition fluxes and their spatial distribution.

2. Since the approach used by EPA relies on model predicted data due to lack of measurements of dry deposition, it is essential to show that the CMAQ model used in the analysis perform adequately when compared against measurement data. The model evaluation presented in Appendix 1 is inadequate for a variety of reasons:

    a. Evaluation is shown for annual averaged quantities that can mask model performance issues, as the compensating effects can cancel out biases and errors in the model. It is a general practice in regulatory applications to choose averaging period commensurate with the measured quantities (e.g., see Morris et al., 2006; Tesche et al., 2006) when evaluating model performance. Therefore, EPA should base model performance statistics using daily or weekly averaged quantities, as appropriate, instead of the annual average quantities. In addition, the statistics should be aggregated for each season, as appropriate.

    b. There is no reason to use normalized mean bias statistics when using annual average quantities. It is recommended to use mean normalized bias for longer averaging periods and fractional normalized bias when using averaging periods of one week or less.

    c. The report does not include a model evaluation performed for NOx purportedly due to lack of available data. However, NO_2 data are available for a model performance evaluation of NO_2. Notwithstanding the issues with the current FRM method for NO_2, it is appropriate to show these model performance results.

       In addition, there are *continuous, highly sensitive and highly precise* measurements of nitric oxide, nitrogen dioxide, total oxidized nitrogen (NOy), and nitric acid available for the SEARCH network in the southeastern U.S.

(Hansen et al, 2003; Edgerton et al. 2006) that could have been relied on to conduct a regional evaluation. Although limited in scope to the Southeast, conducting an evaluation using SEARCH data is better than not conducting any evaluation for NOx at all.

Because of the limited model evaluation shown in the document, the statement on Page 1-5 (Lines 2 to 4) of Appendix 1 that "the model performance results give us confidence that our applications provide a scientifically credible approach for the purposes of this assessment" cannot be supported. Since there is no way to measure the model performance for dry deposition, the estimates of total deposition that rely on predicted dry deposition are highly uncertain.

3. Figure 3.2-1 does not mention or include lightning NOx (LNOx) and soil NOx emissions, so it can be assumed that either those emissions were ignored or were assumed to be negligible (<1% of total). Hudman et al. (2007) show that these emissions can be significant (9% and 27% of total U.S. NOx emissions from soil and lightning, respectively for July 1 to August 2004 period). The magnitude of the lightning source relative to the anthropogenic source in summer 2004 was constrained by the extensive aircraft observations of NOx as part of the ICARTT campaign, so there is high degree of confidence in those estimates. Although these emissions estimates may still have uncertainty associated with them, one cannot ignore a combined source that could be as much as 1/3 of the total NOx emissions over a significant period, especially when estimating regional deposition loads. Additional analysis (Lee Murray and Daniel Jacob, Personal Communications) shows than on an annual basis, lightning and soil NOx may contribute up to 20% of total NOx emissions in the U.S. The exclusion of these emissions used in the CMAQ model simulation further reduces confidence in the modeling results shown in the REA.

   Harvard has conducted preliminary work (Lee Murray and Daniel Jacob, Personal Communications) on estimating contribution of LNOx to nitrogen deposition over several years using the GEOS-Chem model. The results show that LNOx alone (not including the contribution from soil NOx) can contribute ~15% of total annual oxidized nitrogen deposition over the U.S. Another interesting result of their work is that dry deposition accounts for 2/3 of total nitrogen deposition, a result that is different from the results shown in the REA. This indicates that using a single model for a single year to estimate dry deposition could give unreliable estimates. A better approach would be to use multiple models for multiple years.

4. Chapter 3 discusses various uncertainties associated with the analysis, but there is no attempt made to quantify any of those uncertainties. It may not be possible to quantify every source of uncertainty, but there are cases for which this is possible. For example, it is known that ammonia emissions are highly uncertain and studies have been done by EPA to "correct" biases in ammonia emissions using inverse modeling (Gilliland et al., 2003). It is recommended that EPA rerun the CMAQ model with the revised ammonia emissions to quantify the effect of that uncertainty.

5. The REA is not consistent in its use of definitions for NOx and NOy. The first paragraph of Page 1-10 states that “oxides of nitrogen” and “nitrogen oxides” used in this document refers to all forms of oxidized nitrogen compounds, similar to the term NOy used in the scientific community. It is not clear from the document whether the term NOx used in the document refers to (NO + NO_2) or to NOy and it seems to be used interchangeable throughout the document (e.g., in Line 1 of Page 1-14, “NOx” seems to be referring to NOy, but on page 3-3 when discussing emissions, “NOx” seems to referring to sum of NO and NO_2). A better clarification needs to be made in Chapter 1 and followed throughout the document. The following instances of inconsistencies are noted as examples:

    a. Page ES-11: Line 1 refers to annual average NOx concentrations shown in Figure ES-5, but the caption for Figure ES-5 refers to annual average NOy.
    b. Similarly, Page 3-11, Line 27 refers to annual average NOx concentrations, but the caption for Figure 3.2-6 refers to annual average NOy. It is not clear what quantity is shown in the figure.
    c. Page 3-21: The policy-relevant background concentrations are shown for NO_2. If the indicator is NOx, shouldn’t those concentrations be shown for NOx?

Minor Comments:

1. Page 3-29, Line 19: “These findings show that NOx emissions are much higher than NH_3 emissions in most areas of the country”. There is a logical flaw here. The finding that oxidized nitrogen deposition is higher than the reduced nitrogen deposition is a result of emissions that are input into the model, not the other way around.

References:

- Edgerton, E.S., Hartsell, B.E., Saylor, R.D., Jansen, J.J., Hansen, D.A., and Hidy, G.M. (2006). The Southeastern Aerosol Research and Characterization Study: Part III. Continuous Measurements of $PM_{2.5}$ Mass and Composition. *J. Air Waste Mange. Assoc.* **56**, 1325-1341.
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- Morris, R. E. , Koo, B., Guenther, A., Yarwood, G., McNally, D., Tesche, T. W., Tonnesen, G., Boylan, J., and Brewer, P. (2006). Model sensitivity evaluation for organic

carbon using two multi-pollutant air quality models that simulate regional haze in the southeastern United States. *Atmos. Environ.* **40** (26) 4960-4972

- Tesche, T. W., Morris, R. E., Tonnesen, G., McNally, D., Boylan, J., and Brewer, P. (2006). CMAQ/CAMx annual 2002 performance evaluation over the eastern US. *Atmos. Environ.* **40** (26) 4906-4919.

Dr. Myron J. Mitchell

General Comments and Responses to the Charge to the CASAC NO_x/SO_x Secondary Review Panel
Final: July 28, 2009

Comments are provided in *italics*

Charge to the CASAC NOx/Sox Secondary Review Panel

Within each of the main sections of the second daft Risk and Exposure Assessment document, we ask the panel to address the following questions, taking into consideration the changes and additions since the first draft Risk and Exposure Assessment:

Executive Summary:

1. In response to the Panel's review of the first draft Risk and Exposure Assessment, we have included an executive summary of this document. Does the Executive Summary adequately summarize and characterize the key issues driving this review as well as the important findings of the analyses? Does the Panel have any suggestions for clarification or refinement of the Executive Summary?

The Executive Summary does a good job of providing a summarization of the document. The summary, however, needs attention to detail, clarity and consistency. The figures and tables need to have clear and accurate labels. There is some redundancy in the document such as the mentioning at various locations of the importance of looking at the effects of total reactive nitrogen versus NO_x. I know that there has been considerable discussion of the importance of using total reactive nitrogen in these analyses, but some of this usage and other areas of duplication should be reduced. A succinct statement indicating why total reactive nitrogen is the most appropriate metric for nitrogen atmospheric deposition should be included. There needs to be more consistency in the use of the past and present tense. If the focus is on the summary of findings, the past tense is appropriate. However, if emphasis is on the current conditions, the present tense should be used. A clearer transition and better linkage between Section 2.0 (OVERVIEW OF RISK AND EXPOSURE ASSESSMENT) and previous text need to be provided.

In my detailed comments, I have provided a number of editorial corrections and suggestions. There should be consistency of whether or not to use direct references in the executive summary. I would suggest, unless there is a very compelling reason, that direct literature references should not be part of the Executive Summary.

There were no specific questions directed at Chapter 2, but the comments provided for the Executive Summary are relevant to this chapter. Much of the contents of this chapter is based upon further elaboration and justification of the use of ecosystem services in this assessment.

The entire document could be improved by attention to repetition both within and between chapters. A more succinct narrative would provide a more focused presentation of the important issues.

Air Quality Analyses (Chapter 3):

1. This chapter describes an approach for characterizing the spatial and temporal patterns of nitrogen and sulfur deposition in the case study locations including both oxidized and reduced nitrogen, and both wet and dry deposition of oxidized nitrogen, reduced nitrogen, and sulfur. Are the uncertainties associated with these analyses appropriately identified and described?

The Chapter does a good job of describing the spatial and temporal patterns of nitrogen and sulfur deposition including the areas of the case studies. The report needs to further emphasize the inherent limitations associated with estimates of dry deposition. In the Chapter it is sometimes indicated that "measured deposition" is provided. Although good measurements of wet deposition can be provided, there is considerable uncertainty in the dry deposition ***estimates*** *and the importance of this uncertainty should be emphasized in this chapter.*

2. In response to CASAC's recommendation, the RSM analysis presented in the first draft Risk and Exposure Assessment was replaced by an analysis of results from a new series of CMAQ simulations designed to explore the relative contributions of NOx and NH3 emissions to total, reduced and oxidized nitrogen deposition and the relative contribution of SO2 emissions to sulfur deposition. Does this approach enable us to adequately examine the contribution of NOx to total nitrogen deposition?

The relative role of NO_x *and* NH_3 *simulations is better developed in the current document.*

3. The CMAQ application and model performance evaluation is presented in Appendix 1, as recommended by the Panel. Is this analysis sufficient to support the use of the model in this review?

There is more information provided on the CMAQ application and model performance. The comparisons of the model with other independent measures (e.g., CMAQ wet deposition versus NADP/NTN wet deposition estimates) provide important information with respect to the estimates of deposition. Some further comparisons with other deposition measurements would strengthen the report. These results need to be used in showing potential uses and limitations of the model predictions both with respect to regional coverage and temporal changes. These comparisons could include more quantitative analyses of model versus measured performances including comparisons among regions. A clear recognition should be provided that indicates limitations (absence of long-term historical simulations) as well as opportunities (direct linkages between emissions and deposition) in CMAQ modeling. It would be helpful to include some type of critique on the model application results in Appendices 1 and 2 and how these results affect the setting of standards.

Case Study Analyses (Chapters 4 & 5)

Questions related to the individual case study analyses are presented below. Overarching questions across all the case studies include:

1. Are uncertainties appropriately characterized across the case studies? Is there adequate information to allow us to weigh the relative strengths of each case study to inform the standard setting process?

There is sufficient information related to understanding some of the variation associated with each case study. However, it is difficult to translate this uncertainty to developing standards that can be used that include larger regions. Some of this uncertainty is related to the case studies not representing the full spectrum of effects associated with the deposition of nitrogen and sulfur. The tradeoffs in using case studies that focus on sensitive areas versus areas that are spatially representative needs to be stated explicitly. This selection has a major impact on how the results can be extrapolated to other areas.

2. In using the Risk and Exposure Assessment to inform the policy assessment, we plan to focus on aquatic acidification as the basis for an alternative multi-pollutant secondary standard as this is the area where we have the most confidence in our ability to characterize adverse effects. Does the Panel agree with this approach?

The focus on aquatic acidification is reasonable based upon the current information related to both acidification and nutrient enrichment. However, recent findings are suggesting that the recovery of soils from acidification may take an extended period and over the long-term this will also affect the rate of recovery of aquatic ecosystems.

Acidification:

1. Section 4.2 and Appendix 4 describe the analyses used to evaluate the effect of aquatic acidification. The analysis evaluates the ANC in selected lakes and streams in the Adirondacks and Shenandoah relative to three potential ANC cutoff levels (20, 50, and 100 ueq/L) to determine the impact of current levels of deposition in these areas as well as a larger assessment area. Is this data adequate to establish critical loads of deposition for the case study area?

2. The ecological effect function for aquatic acidification (section 4.2.7) attempts to characterize the relationship between deposition and ANC. In order to estimate the amount of NOx and Sox deposition that will maintain an ANC level above a given limit requires the knowledge of the average catchment flux of base cation from weathering of soils and bedrock (i.e., preindustrial cation flux (BC0)). How might we generalize from location specific inputs (F-factor approach) to using this approach on a broader scale - watershed, regionally, or some other way - to generalize beyond individual locations? What other methods should be examined for estimating catchment weathering rates nationwide for surface acidity?

This section now does a better job of describing the importance of mobile anions with respect to soil acidification and resultant effects on the depletion of nutrient cations. Many of the figures in this section need to be redone so that legends, axis values, etc. are easier to read.

This section relies substantially on MAGIC model simulations to show various spatial and temporal trends. It needs to be made clear which version of MAGIC is being used in these calculations since there are major differences with respect to the ability of the model to predict nitrogen watershed chemistry.

In this chapter the term "acidifying deposition" is used. I assume this is done to account for the role of ammonium inputs that can be nitrified resulting in acidification. However, this is not common terminology in the public policy and scientific literature and it may be preferable to use the more standard term "acidic deposition" throughout the document to avoid confusion. On the other hand, if this term is considered to be of importance than it must be clearly defined and justified early in the document.

Within the chapter the term "natural acidity" is used (e.g., pages 4-34 through 4-35). In some of the discussion related to ecological effects of acidification, it is not clear what are the consequences associated with some of the assumptions such as the soils not being a sink for sulfur. We know that some soils are strong sulfur sinks and also there is considerable information that there is a net loss of sulfur from soils (e.g., soils as sulfur sources) (e.g., page 4-37). Such internal elemental cycling can have major effects on the consumption and/or generation of acidity.

In the section on uncertainty and variability (4.2.8) a variety of approaches are provided and these appear mostly to be associated with variation in parameter estimates and how this affects model output. Some discussion on the implicit limitations of the model used (e.g., processes not covered, appropriateness of scale both spatially and temporally, etc.) would help place this section in a broader context of the validity of the model results and any possible major limitations.

The section on ecosystem services is a good summary of helpful information related to ecosystem services and acidification issues (4.3.1.3). It is useful that the report explicitly states the problems of estimating directly how ecosystem services are affected by terrestrial and aquatic acidification.

3. Section 4.3 and Appendix 5 describe the analyses used to evaluate the effect of terrestrial acidification. This analysis uses the Simple Mass Balance Model to determine the impact of current deposition levels on Bc/Al levels relation to three potential Bc/Al cutoff levels (0.6, 1.2, and 10.0) for sugar maple in the Kane Experimental Forest and red spruce in the Hubbard Brook Experimental Forest and a larger assessment area based on the FIA database for 17 states. Is this approach adequate to develop critical loads of deposition for the broader terrestrial acidification case study area? Is the regression analysis between Bc/Al rations and tree health sufficiently described and are uncertainties adequately characterized?

In the sections looking at critical load calculations, there is considerable emphasis on the CMAQ model and its application and other issues related to deposition. I am not sure that there is sufficient balance between these issues and the other issues related to the within system elemental cycles including those processes affecting acidification.

The extrapolation of the critical load calculations for sugar maple and red spruce to other regions (e.g., different states) beyond those of the case studies helped place these results in a broader geographical context. There are some potential issues in looking at these results on state by state basis since these boundaries do not reflect the important characteristics that affect critical loads, but having this information on a state by state basis might be of more interest to policy makers. It is a surprising result that such a high percentage of sites have been compromised with the acidifying total nitrogen and sulfur deposition in 2002 (page 4-62).

In this section there is some use of average critical loads related to three levels of projection. It is not clear if this "average" is meaningful in the context of how critical loads may be applied since the critical load is dependent on specific edaphic features of an area. Does the average take into account how the spatial distribution of edaphic features? Does this "average" apply to specific case study areas or to larger regions? The discussion and analyses that show how specific factors such as parent soil properties affect critical loads calculation is most important.

Nutrient Enrichment:

1. Section 5.2 and Appendix 6 describe the analyses used to evaluate the effect of aquatic nutrient enrichment. The analysis uses the SPARROW model on one stream reach (Potomac River and Neuse River) to determine the impact of atmospheric total nitrogen deposition on the eutrophication index for the estuary. Does the Panel think that the model is adequately described and appropriately applied?

In discussing nutrient enrichment it would be helpful to provide some background on nitrogen being a limiting nutrient in freshwater aquatic systems. Recent evidence suggests that nitrogen limitation is more common than once thought for freshwater systems. Historically, the importance of phosphorus limitation has been emphasized, but more recent work has suggested that nitrogen limitation was less noted due to it being at relatively high levels in many freshwater systems. A major challenge in developing protocols for returning systems to a level of lower nutrient enrichment is defining what attributes and their specific values that are the goals in the restoration of ecosystem type and function. The presentation of areas related to sensitivity to nitrogen loading needs some further clarification on the importance of those areas in the west (e.g., Rocky Mountains of Colorado) that are impacted by relatively low levels of atmospheric nitrogen inputs. Such areas have very different attributes and issues compared to the estuaries that are affected by relatively high levels of anthropogenic nitrogen. There are different issues associated with nitrogen deposition in areas with recent histories of relatively low nitrogen deposition versus areas that are being subjected to very high loadings. How these issues vary among the regions needs to be clearly identified.

The discussion of the case studies related to the Potomac River/Potomac Estuary Case Study Area and Neuse River/Neuse River Estuary Case Study Area including the application of the

SPARROW model is adequate. The broader extrapolation of these results is also helpful. Some of the details provided in this chapter should possibly put into the appendices. The discussion of the uncertainty estimates provides important information on the application of the model simulations. A summary of which of these components is most important with respect to policy recommendations would help clarify what are the most important issues that could affect the interpretation of these results.

2. Section 5.3 and Appendix 7 describe the analyses used to evaluate the effect of terrestrial nutrient enrichment. This qualitative analysis describes the impacts due to nitrogen deposition on the Coastal Sage Scrub community in California and in mixed conifer forests in the San Bernardino and Sierra Nevada Mountains and larger areas where possible. In addition, the effects of nitrogen deposition in the Rocky Mountain National Park supplemental case study location are summarized. How would the Panel apply the threshold values presented in this case study to allow for a broader geographic application that accounts for regional variability? Have the associated uncertainties been adequately characterized?

The emphasis on the California coastal sage scrub (CSS) and San Bernardino Mountains mixed conifer forest (MCF) systems seems appropriate due to their importance with respect to population centers and interactions with nitrogen deposition with other environmental issues including fire susceptibility. Also, other environmental issues such as the potential effects on biodiversity and threatened species is important for these case study areas. The document clearly indicates that although qualitative interactions among these various environmental concerns are well documented there is not good information on the actual quantitative relationships including direct and indirect effects of sulfur and nitrogen deposition. It is noteworthy that the alpine ecosystems of the Rocky Mountains was considered as a case study area, but was not selected. It is suggested that "the ecological benchmarks suggested for alpine ecosystems were comparable to the benchmarks from CSS and MCF ecosystems"(p. 5-57). I am not sure this is true since the alpine systems seem to be especially sensitive to low levels of nitrogen deposition. These is some extensive discussion of the specific issues related to the Eastern Slope of the Rocky Mountains (including Rocky Mountain National Park) in Section 5.3.6.2.

The section on "Uncertainty and Variability" (5.3.8) does a good job of summarizing some of the major issues with a particular focus on the CSS and MCF case studies. On the other hand the "Conclusions" (5.4) section is too short to be very helpful in linking this Chapter with the entire focus of the document.

Additional Effects (Chapter 6):

1. In this chapter, we have presented results from some qualitative analyses for additional effects including visibility, climate and materials, the interactions between sulfur and methylmercury production, nitrous oxide effects on climate, nitrogen addition effects on primary productivity and biogenic greenhouse gas fluxes, and phytotoxic effects on plants. 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?

The section on methyl mercury formation and relationships to sulfur are generally adequate and well done. Some of the wording needs to be changed so that it is clear that this process can occur in areas beyond just surface waters.

The other sections including 6.3 NITROUS OXIDE, 6.4 NITROGEN ADDITION EFFECTS ON PRIMARY PRODUCTIVITY AND BIOGENIC GREENHOUSE GAS FLUXES (including subsections: 6.4.1 Effects on Primary Productivity and Carbon Budgeting; 6.4.2 Biogenic Emissions of Nitrous Oxide; 6.4.3 Methane Emissions and Uptake; 6.4.4 Emission Factors; 6.4.5 Uncertainty), 6.5 DIRECT PHYTOTOXIC EFFECTS OF GASEOUS SO_X *AND* NO_X *[including subsections: 6.5.1* SO_2*; 6.5.2 NO,* NO_2 *and Peroxyacetyl Nitrate (PAN),6.5.3 Nitric Acid (*HNO_3*)] are adequate in the context of the needs of the current report.*

Synthesis of Case Studies (Chapter 7):

1. Here, the case study analyses are integrated and synthesized within the conceptual framework of ecosystem services as shown in Figure 7-2. Where possible, we have quantified select ecosystem services associated with the ecological effects targeted in this review. This chapter discusses adversity by characterizing the degree to which ecological effects are occurring under given levels of deposition to inform the discussion of adversity in the policy assessment and standard setting process. To what extent do you think the description of ecosystem services provides a useful framework in the case study analyses for informing standard setting? Does the Panel have suggestions for additional consideration or characterizations for ecosystem services related to the case studies?

The information in Chapter 7 provides a summary of previous information including emphasis on ecosystem services. Much of this information has been previously stated in earlier chapters, but having this summarized information in a single chapter is helpful. It should be emphasized that the results from these case studies can only be regionally extrapolated to those areas of similar characteristics with respect to sensitive to sulfur and nitrogen deposition.

2. Based on the information presented in the current Risk and Exposure Assessment, given adequate time and resources, is there enough information to inform setting separate standards based on the other targeted ecological effects, specifically, terrestrial acidification, aquatic nutrient enrichment, and terrestrial nutrient enrichment? If not, how can our understanding of these ecological effects be enhanced in time to inform the next 5-year review?

There appears to be sufficient information provided in the current "Risk and Exposure Assessment" given additional time and resources to form standards with respect to effects of acidification and nutrient enrichment on terrestrial and aquatic systems. Clearly, however, there is considerable uncertainty on these effects especially with respect to both the current and future deposition of sulfur and nitrogen. Similarly there are complexities associated with interactions with a broad range of factors including land use, climate, effects of invasive species, etc. that result in uncertainties for predicting the effects of sulfur and nitrogen deposition. It may be helpful to show the range of ecosystem services and what is known about how they are affected by acidification and nutrient alteration due to sulfur and nitrogen deposition. It would be helpful to show what is known and unknown about the linkages over a range of ecosystem *services. Inclusion of what is needed to provide information in the future about the effects of nitrogen and sulfur deposition on various ecosystem services would be helpful for both the current REA and also setting the stage for other efforts to evaluate multiple pollutant/environmental effects on these ecosystem services.*

Detailed Comments and Responses to the Charge to the CASAC NO_x/SO_x Secondary Review Panel Final: July 28, 2009

Page	Line(s)	Comment
xxiv	4-5	This statement makes no sense: "Ecosystem Structure: Refers to the species composition, distribution, and interactions with some abiotic attributes of the environment s they vary through space and time".
xxiv	21	Change "*This* indicator may either be the actual criteria air pollutant" to "*An* indicator may either be the actual criteria air pollutant".
xxv	2	Delete "As a result".
xxvii	3	Delete "reduced" twice in this line. Why use the term reduced?
xxvii	3	Give the charge for "NO_2" (nitrite) as "-".
xxviii	7	Change to "other *forms of* precipitation".
ES-2	12-13	Change "slower biomass growth" to "lower rates of production".
ES-2	14	Change "In addition to acidification, NO_x acts" to "In addition to contributing to acidification, NO_x acts".
ES-2	22-23	Change "the ecosystem is receiving more nitrogen than it uses" to "an ecosystem is receiving nitrogen in excess of biotic nutritional needs".
ES-2	23	Delete "also".
ES-2	24	Clarify what "This" refers to. Does this mean nitrogen deposition, primary productivity and/or terrestrial carbon cycling?
ES-2	26	Change to "Lichens".
ES-2	27-28	This statement seems out of place. Does this statement have any relevance to lichens? Clarify what aspects of biodiversity have been reduced in grasslands.
ES-3		Figure ES-1. Within the figure change "Soil Process" to Soil solute generation".
ES-4	9	Change "to determining when the" to "to determining when *and where* the"

ES-5	8	Replace “enrich” with “impact”.
ES-5	8	Clarify what “this” refers to.
ES-5	12	Change “while the Ecological Effect Function (box 6) relates the deposition metric into the” to “while the Ecological Effect Function (box 6) links the deposition metric to the relevant”.
ES-5	19	Change “the degradation of” to “deleterious affecting”.
ES-6	2	Change “Because ecosystems are diverse” to “Because ecosystems differ”.
ES-7		In Table ES-1 the Adirondack Mountains should be referred to as the “Adirondacks” and not the Adirondack”.
ES-8	9	Change “ecosystem services is being used as an umbrella term” to “the term ecosystem services is being used as a broad concept”.
ES-8	10	Change “It is a way to help explain” to “The evaluation of ecosystem services helps to explain”.
ES-9	3-4	Change “some of the ecosystem services likely to be affected are readily identified, while others will remain unidentified” to “ only some of the ecosystem services that are likely to be affected can be readily identified”.
ES-9	4-6	Change “Of those ecosystem services that are identified, some changes can be quantified, whereas others will remain unidentified” to “Of those ecosystem services that are identified, only subset of changes will likely be quantifiable”.
ES-9	6-7	Change “Within those services whose changes are quantified, only a few will likely be monetized, and many will remain unmonetized” to “For those quantifiable services only a few will be subject to monetization”.
ES-9	8	Change “A conceptual model integrating” to “An example of a conceptual model of effects on aquatic ecosystems is used to integrate”.
ES-9	12	Change “can be used to inform a policy judgment” to “can be used to in developing policy”
ES-9	16	Change “inform” to “provide”.
ES-9	18-21	Figure caption needs to be changed to indicate that this is an example focusing on aquatic ecosystems.

ES-11 3	The term "magnitude" can be misleading. The correspondence between the actual amount and relative spatial patterns of measured versus modeled concentrations needs clarification.
ES-12 7	Change "information about meteorology and land use in each grid cell of the domain" to "information about meteorology and land use both of which are critical components in affecting dry deposition".
ES-13 5-7	Change "In the East, high levels of deposition exceeding 18 kg S/ha/yr occur in the immediate vicinity of isolated major sources, as well as in and near areas having a high concentration of SO_2 sources" to "In the East, the highest levels of deposition that exceeding 18 kg S/ha/yr occur in proximity to sources of high SO_2 emission" .
ES-15 13	Change "Acidification can degrade the health of terrestrial and aquatic ecosystems" to "Acidification can have deleterious impacts on terrestrial and aquatic ecosystems".
ES-15 19	Delete "method".
ES-15 28	Delete "the additional".
ES-16 12-13	Change "direct relationship between ANC and fish and phyto-zooplankton diversity and abundance" to "direct relationship between ANC and the diversity and abundance of fish and phyto-zooplankton".
ES-16 13	Change "MAGIC" to "The MAGIC model".
ES-16	Within this page direct citations are provided. This does not seem to be consistent with other portions of the Executive Summary. I would suggest that these should be deleted for consistency. This problem is also found in other parts of the Executive Summary.
ES-17 9	Change "Calcium and Al are strongly" to "Calcium and Al concentrations are strongly".
ES-17 13	See previous comments on the use of citations in the Executive Summary.
ES-17 18	Change "The tree species most sensitive" to "Tree species sensitive"
ES-17 19	Delete "a deciduous tree species".
ES-17 20	Delete "a coniferous tree species".
ES-17 22	Change "to" to "on".

ES-17	23	Delete “both”.
ES-18	6	Change “total removal” to “total harvest”.
ES-18	6	Remove “from timberland”.
ES-18	9	Change “roughly” to “approximately”.
ES-18	10	Remove “from timberland”.
ES-18	11-12	Change “spruce forests are home t o the spruce-fir moss spider (endangered), the rock gnome lichen 12 (endangered), and the Virginia northern flying squirrel (delisted, but considered important).” to “spruce forests are important habitats for endangered species including the spruce-fir moss spider, the rock gnome lichen, and the Virginia northern flying squirrel (delisted, but still considered important).
ES-19	1-4	Change “Some organisms may at first respond positively to an initial increase in nutrients, exhibiting an increase in growth due to fertilization effects. However, as the nutrient load continues to rise, the imbalance can have negative effects either in the organism’s response or in the invasion of new organisms that benefit from increased nutrients” to “ Some organisms may at first respond to an increase in nutrients with increased growth. However, as nutrient load continues to rise, the resulting imbalance can have negative effects either directly on the organism or indirectly by the invasion of other species that are better competitors under high nutrient conditions”.
ES-19	12-14	Change “Nitrogen is an essential nutrient for aquatic ecosystem fertility, including lake, marine, and estuarine ecosystems, and is often the limiting nutrient for growth and reproduction in many of these ecosystems” to “Nitrogen is often a limiting nutrient for lake, marine, and estuarine ecosystems”.
ES-19	15	Delete “of a system”.
ES-19	21	Change to “nitrogen *enrichment* now represents”.
ES-19	26	Change “Due to the cascading impacts and effects of nitrogen enrichment” to “Due to the cascading impacts of nitrogen pollutants”.
ES-20	2	Change “estimation” to “estimate”.
ES-20	5	Change “In this assessment” to “In the current assessment”.
ES-20	14	Change “reductions in additional” to “reductions from additional”.

ES-20	15	Change "resident commercial species" to "resident commercial species important for various fisheries".
ES-20	24-25	See previous statements about citations in the Executive Summary.
ES-20	29	See previous statements about citations in the Executive Summary.
ES-20	29	Change "that was only about" to "of only ~".
ES-20	31	See previous statements about citations in the Executive Summary.
ES-21	2	Change "only source of nitrogen to these systems" to "the dominant source of nitrogen to these systems".
ES-21	5-6	Change "; creating increased growth rates in some species over others, which changes competitive interactions among species; and nutrient imbalances" to ". This higher N availability affects the relative interspecific competitive of plant species resulting in changes in species composition and vegetation structure".
ES-21	13-14	Change "to cause increased litter accumulation in the soils and carbon storage in aboveground biomass" to increased carbon storage in aboveground biomass and litter".
ES-21	16	Change "can" to "may".
ES-21	17	Change "by nitrogen limitation can now better compete and alter species dominance" to "by nitrogen limitation are more competitive".
ES-21	19-20	Change "the leaching of NO_3^- in soil drainage waters" to "soil NO_3^- leaching".
ES-21	20	Change "in stream water" to "in surface waters".
ES-21	22-23	Delete "; however, these measurements are not always widely available".
ES-21	26	Change "that nitrogen" to "that increased nitrogen inputs".
ES-21	28-	Is the description on the "extent of ecosystems" or the extent of the ecosystems impacted by nitrogen deposition?
ES-22	3-6	Delete these lines.
ES-22	19-23	This seems like a rather detailed finding and could be deleted from the Executive Summary.

ES-23 8	Change “could be quite high” to “is quite high”.
ES-23 11-13	Change “enrichment potentially include decline in CSS habitat, decline in protection of native species, increase in abundance of nonnative grasses, and increase in wildfires” to “enrichment potentially include declines in CSS habitat the protection of native species, and increases in nonnative grasses and wildfires”.
ES-23 16	Change “helps regulate” to helps control”.
ES-23 17	Change “upset” to “disrupted”.
ES-23 19	Change “could” to “may”.
ES-24 1	Change “SO_x deposition on methylmercury production” to “SO_x deposition and resultant change in soil and wetland SO_4^{2-} concentrations in affecting methylmercury production”.
ES-24 3	Change “scope of this review” to “scope of the current review”.
ES-24 9-10	Change “While there are many uncertainties associated with these analyses, from a scientific perspective there is confidence that known or anticipated adverse ecological effects are occurring” to “Although uncertainties exist, there is strong evidence that known or anticipated adverse ecological effects are occurring”.
ES-24 12-13	Change “Of all the case study analyses, there is most confidence in the ecological responses, effects, and benefits associated with aquatic acidification” to “Within the case study analyses, there is most confidence in the ecological responses, effects, and the deleterious impacts associated with acidic deposition”.
ES-24 13-14	Change “and there is a fair amount of confidence about those associated with terrestrial acidification” to “Similarly, the importance associated with the impacts of acidic deposition on terrestrial systems is clearly documented”.
ES-24 15	Change “benefits” to “deleterious impacts”.
ES-24 18	Delete “However”.
ES-24 20	Change “only” to “dominant”.
ES-24 22	Change “benefits” to “deleterious impacts”.

ES-24	25-26	Change “terrestrial acidification may be the most useful in terms of developing a secondary” to “terrestrial acidification should be most useful in developing a secondary”.
ES-25		These specific citations should be removed f rom the Executive Summary.
1-2	2	Change “The species of nitrogen and sulfur” to “The chemical species of nitrogen and sulfur’.
1-2	5-7	Change “because NO_x, SO_x, and their associated transformation products are linked from an atmospheric chemistry perspective, as well as from an environmental effects perspective” to “because the atmospheric chemistry and environmental effects of NO_x, SO_x, and their associated transformation products are linked”.
1-2	10	Change “of these two pollutants has been conducted” to “of SO_x and NO_x as well as total reactive N has been conducted”.
1-2	11	Delete “at this time”.
1-2	16-17	Change “in an ecologically meaningful way” to “that is ecologically meaningful”.
1-3	1	Change “see” to “go to:”.
1-4	13	Why “identical”?
1-4	22	Change “This draft document” to This latter draft document”.
1-5	29	Change “At that time, EPA was aware that SO_x have” to “At that time, EPA was aware that SO_x has”.
1-6	1	Change “specific SO_x concentrations” to “specific atmospheric SO_x concentrations”.
1-8	8	Change “at that time” at the time of the report (1995).
1-8	14	Change “particular relevance to this review” to “particular relevance to the current review”.
1-10	13-20	Should some mention be made of organic forms of N in the atmosphere including DON? It is noteworthy that in Figure 1.3-1 (page 1-11) that organic forms of N are shown
1-11	9-15	Although the figure shows some of the organic atmospheric S forms. There is no mention of these chemical species in the text.

1-12	19	Change “not high enough” to “not sufficiently high”.
1-13	6-7	Change “Both are essential and sometimes limiting, nutrients needed for growth and productivity” to “Both N and S are essential macronutrients”.
1-13	7	Change “Excess” to However, excess”.
1-13	10	Change from “These effects include slower growth” to “These effects include slower biotic growth”.
1-13	23	Change “Models suggest that” to “Models for the latter study area suggest that”.
1-13	28	Change “acidification effects from acidifying deposition” to “acidification effects from atmospheric deposition”.
1-14	9	Change “that leads” to “that may lead”.
1-14	21	Change “quality in the western United States (U.S. EPA, 2008, Section 3.3" to “quality in the western United States, a region especially sensitive to increased nitrogen atmospheric inputs (U.S. EPA, 2008, Section 3.3"
1-14	23	Change “which leads to eutrophication” to “which may lead to eutrophication”.
1-14	27	Change “in highly eutrophic estuaries” to “in some eutrophic estuaries”.
1-14	30-31	Change “In terrestrial ecosystems, there are multiple chemical indicators for the alteration of the biogeochemical cycling of nitrogen that is caused by total reactive nitrogen deposition” to “In terrestrial ecosystems, there are multiple chemical indicators that the biogeochemical cycling of nitrogen has been altered by the deposition of total reactive nitrogen”.
1-14-15		Change “Nitrate leaching” to “Nitrate leaching from terrestrial ecosystems”.
1-15	2	Change “the onset of leaching” to “the atmospheric deposition threshold for nitrate leaching”.
1-15	7	Change “occurring at 3 kg” to “occurring at atmospheric inputs as low as 3 kg”.
1-15	13	Change “this” to “the current”.

1-15	19	Change “In watersheds where changes in sulfate deposition did not produced an effect” to “In watersheds where changes in sulfate deposition did not result in changes in methylmercury generation”.
1-15	20	Change “meaningful” to “substantial”.
1-16		Figure 1.3-3 See previous comments on Figure ES-1 (Is this figure the same as Figure 1.3-3?)
1-17	18-20	This sentence needs to be reworded.
1-18	15-17	This sentence is confusing. What is meant by “the uncertainties in the estimated reductions”? Is the uncertainty on the amount of atmospheric reduction that will occur or uncertainty on the effects of reductions?
1-19	21	Change “Identifying important chemical species in the atmosphere” to “Identifying important N and S chemical species in the atmosphere”.
1-21	6	Change “All of Figure 1.4-1" to “All of the components of Figure 1.4-1".
2-6	14-15	Change “to total loadings of in the environment” to “to the combined atmospheric loadings of both elements”.
2-7	7	Change “a broad look into the” to “an overview of”.
2-7	8	Change “services that is one tool that can help link” to “services. The analysis of the effects on ecosystem services will help link “.
2-7	10-11	Change “In this Risk and Exposure Assessment, ecosystem services is used as an umbrella term to aid in describing the impacts of ecological effects on public welfare and to help explain how” to “In this Risk and Exposure Assessment, ecosystem services is used to show the impacts of ecological effects on public welfare and help explain how” .
2-8	3	Change “data were not abundant enough” to “data were not sufficient”
2-8	19-20	This sentence seems out of place.
2-15	2-3	Does the statement “The analysis of ecosystem services for the aquatic acidification focused on recreational fishing” indicating that focus for the current assessment of the general analysis of ecosystem services in aquatic ecosystems. I believe the sentence should be changed to: “The current assessment the analysis of effects on ecosystem services from aquatic acidification focused on recreational fishing”.
2-15	24	Change “little data is” to “little data are”.

2-16	2	Change "ecosystems are addressed" to "ecosystems were addressed".
3-2	25	Change "species-specific" to chemical species-specific".
3-16	13-24	The term "measured deposition" is used and this needs to be changed to "estimated deposition" since dry deposition are inferred by model calculations and not measured rates. Such nomenclature needs to be changed throughout the document.
3-22	8-16	The title should be changed to "Non-Atmospheric Loadings of Nitrogen and Sulfur". The inclusion of examples associated with the contribution of other sources such as the weathering of sulfur minerals should be considered.
3-27	16	Change "White Face" to "Whiteface".
3-93	23-28	The statement that "Although there are uncertainties in the data, models, and techniques used for this assessment, this analysis relies upon the most applicable measurements and state-of-the-science models. In addition, these data and models are used in a manner that considers their relative strengths and limitations. The inherent uncertainties are not expected to measurably affect the robustness of these conclusions and findings on the characterization of concentrations and deposition" places a very "positive spin" on these model applications, I am not sure that this is entirely justified based upon the level of uncertainties especially with respect to dry deposition estimates of the available models.
4-1	26-27	Change "Under natural conditions (i.e., low atmospheric deposition of nitrogen and sulfur), the limited mobility of anions in the soil controls the rate of base cation leaching" to "Under conditions of low atmospheric deposition of nitrogen and sulfur, the naturally produced bicarbonate anion is often the dominant mobile anion with SO_4^{2-} and NO_3^- playing a limited role with respect to cation leaching".
4-1	27-30	Change "However, acidifying deposition of nitrogen and sulfur species can significantly increase the concentration of anions in the soil, leading to an accelerated rate of base cation leaching, particularly the leaching of Ca^{2+} and Mg^{2+} cations" to "Increased atmospheric deposition of sulfur and nitrogen can result in marked increases in SO_4^{2-} and NO_3^- soil fluxes resulting in the concomitant leaching of nutrient (Ca^{2+}, Mg^{2+}) and toxic (Al^{n+} and H^+) cations".
4-2	8	Change to "Criteria for case study selection".
4-2	11	Change "Current conditions for other areas" to "Current conditions for these other areas".

4-2	26-28	Change "sum of soil and water processes that occur upstream within a watershed, it also reflects the results of watershed-scale terrestrial effects, including nitrogen saturation, forest decline, and soil acidification (Stoddard et al., 2003)." to "sum of terrestrial and aquatic processes that occur upstream within a watershed. Important terrestrial processes include nitrogen saturation, forest decline, and soil acidification (Stoddard et al., 2003)".
4-3	8	Change "certain" to "some".
4-3	10	Change "where strong acids are deposited into the soil" to "where strong mineral acids (e.g., H_2SO_4 and HNO_3) are deposited or generated within the soil.
4-2	13-14	Change "inorganic Al can become mobilized, leading to the leaching of Al into soil waters and surface waters" to "inorganic Al can be mobilized, leading to the leaching of Al from soils to surface waters".
4-2	15	Change "differently" to "differently to acidic deposition".
4-2	15-16	Change "on sensitive species" to "on different ecosystems and species".
4-2	20	Change "migrates" to "leaches".
4-3	23	Change "maintains the balance of electric charge" to "maintains electroneutrality".
4-4	1	Delete "further".
4–4	8	I would disagree that episodic acidification is more important than chronic acidification.
4-4	7-8	Change to "Short-term (i.e., hours or days) episodic changes in water chemistry have perhaps the most significant biological effects" to "Short-term (i.e., hours or days) episodic changes in water chemistry have important biological effects".
4-4	9	Change "rainstorms or snowmelt" to "precipitation or snowmelt events"
4-4	10	Change "which tends to provide less neutralizing of atmospheric acidity as compared with" to "than tends to provide less acid neutralizing than water passing through".
4-4	12	Change "storm runoff or snowmelt" to "events".

4-5	9-10	Change “receptors” to “parameters”.
4-5	13-14	Delete “Although ANC does not relate directly to the health of biota” and start sentence with “The utility”.
4-5	22	Delete “the”.
4-5	24	Change “Low ANC concentrations have” Low ANC has”.
4-5	30	Change “(Figure 4.2-1, a), which” to “(Figure 4.2-1, a) that”.
4-6	3	Change “has been found in studies” to “has been found in various studies”.
4-6	8-9	Change “Below 100 μeq/L, it has been shown that fish fitness and community diversity begin to decline” to “Below 100 μeq/L ANC fish fitness and community diversity begin to decline” .
4-6	11	Delete “decline; however, the overall health of the community remains good”.
4-6	13	Change “that are sensitive to negative effects on biota that are sensitive to acidification” to “that are sensitive to acidification”.
4-7	2	Change “had to have” to “need to have”.
4-7	9	Delete “primarily”.
4-7	17	Change “fishers” to “fisherman”–this may not be a “sex neutral” term, but fishers refers to a type of animal.
4-8	2	Delete “in these states”.
4-8	13-14	Change “services, such as hydrological regime regulation and climate regulation” to “services associated with hydrology and climate”.
4-8	15	Delete “specific”.
4-8	16-17	Change “delicate aquatic food chains” to “aquatic food webs” .
4-8	19	Delete “it is worth noting that”.
4-8	20-22	Delete “For example, these biological control services may serve as “intermediate” inputs that support the production of “final” recreational fishing and other cultural services”.

4-9	4-5	Change “The regions of the United States with low surface water ANC values are the areas that are sensitive to acidifying deposition” to “The regions of the United States with low surface water ANC values are sensitive to acidifying deposition’.
4-9	6-7	Delete “at their existing ambient concentration levels”.
4-9	14	Change “surface water data” to “analyses of sulfur waters”.
4-9	19	Delete “are estimated to”.
4-9	21-24	Change “In 2002, Stoddard et al. (2003) took another comprehensive look at the level of acidification within all of these regions. Although improvement in ANC occurred, about 8% of lakes in the Adirondack Mountains and 6% to 8% of streams in the northern Appalachian Plateau and Ridge/Blue Ridge region were still acidic at base-flow conditions” to “Stoddard et al. (2003) suggested that although improvement in ANC had occurred ~8% of lakes in the Adirondack Mountains and from 6% to 8% of streams in the northern Appalachian Plateau and Ridge/Blue Ridge region were acidic at base-flow conditions”.
4-10	9-12	Change “After considering this information, the Adirondack Mountains and the Shenandoah Mountains (referred to in this chapter as Adirondack and Shenandoah case study areas, respectively) were selected. The rationale for choosing these two case study areas is described in the following subsections” to “Using the rationale described in the following subsections the Adirondack Mountains and Shenandoah Mountains were selected for case study areas”.
4-10	16	Change “The case study area” to This area”.
4-10-11		Delete “, which all draw water from the preserve”.
4-12		For Figure 4.2-3, the axes legends and numbers are too small.
4-13		For Figure 4.2-4, the axes legends and numbers are too small.
4-14	6-7	Delete “because it can no longer be measured”.
4-14	7	Change “Likewise, it is also difficult to determine” to “Likewise, it is also difficult to empirically determine”.
4-14	9	Change “hydrological” to “biogeochemical”–MAGIC is not a hydrologic model.
4-14		10 Change “quality levels” to “chemistry”.

4-16		The insert on critical loads includes the value of 50 $meq/m^2 \cdot yr$. This value may be confusing in using a load based upon charge versus mass since much of the proceeding discussion including inputs used mass values.
4-17	19	Change "the condition" to "the modeled condition".
4-18	5-8	It is important to mention that although SO_4^{2-} still dominates the relative importance of NO_3^- is increasing substantially. Also, comparing concentrations of SO_4^{2-} and NO_3^- in surface waters can be misleading since there may be substantial losses of NO_3^- due to biotic processes in watersheds.
4-18	13	Were these declines in Al statistically significant?
4-18	14	Change "significant" to "substantial".
4-20	1-5	Reword this sentence it makes not sense.
4-20	7	Change "is" to "was".
4-20	10	Change "are" to "were".
4-21	2-5	Change "Percentage of Adirondack Case Study Area lakes in the five classes of acidification (i.e., Acute, Severe, Elevated, Moderate, Low) for years 2006 and 1860 (preacidification) for 44 lakes modeled using MAGIC. Error bar indicates the 95% confidence interval" to "Percentage of Adirondack Case Study Area lakes in the five classes of acidification (i.e., Acute, Severe, Elevated, Moderate, Low) for years 1860 (preacidification) and 2006 for 44 lakes modeled using MAGIC. Error bar indicates the 95% confidence interval". (Make similar changes in other figure captions including 4.2-19)
4-21	8-11	Change "Sites labeled by red or orange dots have less buffering ability than sites labeled with yellow and green dots, and hence, indicate those lakes that are most sensitive to acidifying deposition, due to a host of environmental factors" to "Sites indicated by red or orange circles have less buffering ability than sites labeled with yellow and green circles, and hence, indicate those lakes that are most sensitive to acidifying deposition".
4-22	2-7	In figure caption change "dots" to "circles". *Make similar changes in all figure captions and text.*

4-23	7-9	Change “In considering the future responses of lakes to current emissions and given the current condition of the lakes, the question becomes whether lakes can recover to healthy systems (i.e., ANC > 50 μeq/L)” to “In considering the future responses of lakes, the question becomes whether lakes can recover to healthy systems (i.e., ANC > 50 μeq/L) under current levels of deposition”.
4-24	4-11	Change “Based on a deposition scenario that maintains current emission levels to years 2020and 2050, the simulation forecast indicates no improvement in water quality” to “Based on a deposition scenario that maintains current emission levels to up to years 2020 and 2050, the simulation forecast indicates no improvement in water quality over either of these periods”.
4-24	15	Change from “will likely not improve the acidification of lakes” to “will not likely improve the recovery from acidification”.
4-24	15-17	Delete this sentence.
4-24	24-25	Change “At this time, it is unclear why ANC initially improved and is now declining” to “It is not known what has caused this temporal pattern of ANC in this case study”.
4-25	Table 4.2-4	Indicate what “+/-“ columns signify.
4-26	3	Change “changed statistically” to “did not significantly differ”.
4-26	19	Change “industrially generated acidifying deposition” to “acidic deposition”.
4-26	21	Change “is” to “was”.
4-31	3	Change “Based on a deposition scenario that maintains current emission levels to 2020 and 2050" to Based on a deposition scenario that maintains current emission levels to years 2020 and 2050".
4-31	12	Change “country” to ”U.S.”.
4-31	13	Change “across populations” to “across various populations”.
4-31	14	Change “picked” to “selected”.
4-31	15	Change “to make estimates of regional extent of condition (e.g., number of lakes, length of stream)” to “to make regional estimates of surface water conditions”.

4-31	21	Change “to be susceptible” to “to be especially susceptible”.
4-32	24	Change “area” to “areas”.
4-32	32	I don’t believe the term “ecoregion” been defined in the document.
4-26	19	Change “industrially generated acidifying deposition” to “acidic deposition”.
4-34	10	Change “SO_2" to “SO_x”.
4-34	13-14	Change “One hundred 17 lakes of the 169 lakes modeled for critical loads are part of a subset of 1,842 lakes in the Adirondack Case Study Area” to “ Of the 169 lakes modeled for critical loads, 117 of these lakes were within 1,842 lakes in entire the Adirondack Case Study Area”.
4-34	14-15	Delete “which include all lakes from 0.5 to 2,000 ha in size and at least 1 m in depth”.
4-34	21	Change to “13% *of the total population*”.
4-34	22	Change “some lakes would have never had ANC” to “some lakes would have never had ANC”.
4-34	24-25	Change “estimate based on the critical load alone” to “estimate based solely using the critical load criterion”.
4-34	26	What is meant by “natural”? Does this refer to current conditions or preindustrial concentrations? The term natural with respect to surface water acidity needs to be defined. It might be clearer to discuss the role of DOC in these waterbodies that have historically low ANC.
4-35	20	Change “the same” to “similar”.
4-36	12-14	Change “a host of catchment processes and environmental factors that affect the level of base cations (e.g., Ca^+, Mg^+) concentrations and the sinks of nitrogen and sulfur in the lake and terrestrial catchment” to “a series of biogeochemical processes that produce and consume acidity in watersheds”.
4-36	19-21	Change “Although ANC does not directly affect the health of biotic communities, it ameliorates acidity-related biotic stress that provides an “ecological indicator” of overall integrity of the ecosystem” to “Although ANC has not generally been used as a parameter for predicting the health of biotic communities, it provides useful information of the potential acidity-related biotic stress and hence is a useful “ecological indicator”.

4-36	22	Delete "then".
4-37	9-10	Change "To convert surface water concentrations into surface water fluxes, multiply by runoff (Q) (in m/yr) from the site" to Surface water concentrations are converted to fluxes by multiplying concentrations by runoff (Q) (in m/yr)".
4-37	12	Change "between plants and soil is ignored" to "between plants and soil is negligible".
4-37	19-21	This sentence is confusing. Certainly the nitrogen and sulfur biogeochemical fluxes and transformations affect acidity.
4-40	1-3	This is not just a problem for the United States. The estimate of weathering rates (including the generation of base cations) is a major limitation for many biogeochemical analyses and interpretations
4-42	22	Delete "a coniferous tree species" and " a deciduous tree species".
4-48	6	Delete "and forest".
4-62	14-18	The following statement "Collectively, these results suggest that the health of at least a portion of the sugar maple and red spruce growing in the United States may have been compromised with the acidifying total nitrogen and sulfur deposition in 2002; even with the lowest level of protection, half the states contained sugar maple and red spruce stands that were negatively impacted by acidifying deposition" will receive considerable attention. It is important that any caveats be provided on these results so that the interpretation is placed in the most complete picture of the state of the science.
4-65	10	Change "was" to "is".
4-66	17	Does the "average critical loads" have any real meaning in the context of setting critical loads. I would suggest that the range is the most important and demonstrates and clearly shows how edaphic factors can have a major influence on critical loads.
4-67		Figure 4.3.9 See previous comment with respect of providing the values for average critical loads.
4-68		Figure 4.3.10 See comment above on the use of average critical loads in this figure.

4-69		The discussion of uncertainty is important and highlights some of the issues related to the actual calculation of critical loads. It may be over stretching the uncertainty analyses to suggest that "If all or a large majority of estimates indicate that the critical load of a system is exceeded with current total nitrogen and sulfur deposition rates, the probability is high that deposition is greater than the critical load and that the trees and vegetation in that system are being negatively impacted by acidification". The use of the term "probability" seems out of place and suggests that this approach has a stronger statistical underpinning than is the actual case. The key factor is what are the range of values that affect these calculations and how confident are you in using these values in making these calculations. Similarly the term "certainty" would suggest more confidence in these estimates than may actually be the case.
4-71	3-5	This type of calculation in which it is clearly shown how different values can be obtained for critical loads based upon specific edaphic factors (e.g., parent material acidity) is a useful approach and show how this factor can have a dramatic impact on these calculations.
5-2	24	Change "resulting in increased productivity (e.g., of algae or aquatic plants)" to "resulting in increased primary productivity".
5-2	25	Change "increases, dissolved" to "increases with concomitant increases in organic matter production, dissolved".
5-4	24	Change "for a portion of the nitrogen input" to "for a portion of this nitrogen input".
5-8	21-22	Delete "Therefore, if the susceptibility is known and held constant, a curve can be created".
5-9	10	The determination of on the nitrogen inputs in "pristine" conditions is a difficult task. I agree that providing some upper and lower bounds is a useful approach.
5-10	19	Change "fish" to "seafood". Throughout this section change wording so that is clear that the entire "seafood" resources, not only fish are being considered.
5-11	5	Change "fish" to "seafood".
5-12	10	Change "and reduce" to "and reduced".
5-14	7	Change "are" to "were".
5-14	14	Change "175 million days" to "175 million participant days".

5-14	15	Change “more than 35 million days per year” to “more than 35 million participant days per year”.
5-14	19-20	Change “including climate, biological, and water regulation; pollution detoxification; erosion prevention; and protection against natural hazards” to “including those important for the quality and quantity of water and effects on climate including impacts from storms”.
5-15	13	Change “defining” to “determining”
5-29	3	Change “under suspicion of eutrophication”likely to be subject to eutrophication”.
5-29	31	Not sure if “in prep” information should be cited.
5-47	2	There is a mixture of aquatic and terrestrial ecosystem benchmarks in “Figure 5.3-2. Benchmarks of atmospheric nitrogen deposition for several ecosystem indicators” with the inclusion of the diatom changes in the Rocky Mountain lakes.
5-57	7-8	I am not sure that the following statement is valid: “However, the ecological benchmarks suggested for alpine ecosystems were comparable to the benchmarks from CSS and MCF ecosystems”. At a minimum some supportive statements are needed.
5-64	10-15	The discussion on amounts of nitrogen in throughfall appears to suggest that these differences are due to differences in atmospheric deposition. Although some of this difference may be due to the amounts of atmospheric deposition, the importance of canopy exchange in contributing to nitrogen in throughfall may also be important. See also on page 5-68 with respect to the establishment of critical loads based upon throughfall nitrogen flux.
5-67	1-3	The statement that “increased litter deposition may facilitate faster rates of microbial decomposition initially but may reduce decomposition over the long term because of changes in the C:N ratio and increasing lignin content over time” needs clarification.
5-72	1	Change “classic” to “documented”.
5-75	17	Delete “from nitrogen-saturated forest soils”.
5-75	17	Change “into streams” to “into streams of the northeastern U.S.”.
6-2	31-32	Change “aquatic environments” to “aquatic and terrestrial environments, including wetlands,”

6-2	33	Delete “surface water”.
6-11	1	Change “emissions” to “deposition”.
6-12	24	Change “its global warming potential” to “its global warming potential per molecule”.
6-14	14-15	Change “Nitrogen deposition can affect the patterns of carbon allocation because most growth occurs above ground” to “Nitrogen deposition can affect the patterns of carbon allocation between above and below ground production”.
6-14	15	Change “This increases the shoot-to-root ratio” to “Increased nitrogen availability increases the shoot-to-root ratio”.
6-11	20	Change “Reducing SO_x” to “Reducing SO_x emissions”.

Mr. Rich Poirot

General Comments on Chapter 3
This chapter summarizes the technical approach for characterizing spatial and temporal patterns in the atmospheric deposition of S and N compounds – originating from anthropogenic SO_x, NO_x and NH_x emissions - in the various case study areas. As a practical matter, these estimates are made with relatively high spatial and temporal resolution covering the entire US, and with flexible modeling methods that will allow scaling of estimated deposition patterns and effects to other sensitive areas, and would also support evaluating potential responses to changes in deposition, air quality and/or precursor emissions.

Generally, I think the chapter is clearly written, conveys useful information, and presents convincing support for the approaches taken to provide the best possible estimates of air quality and deposition in the case study areas and elsewhere within the US measurement and modeling domains. One general criticism is that on several occasions the discussion seems unnecessarily qualitative in places where it would take no more space and provide more useful information if quantitative information was provided. Rather than saying "much greater" or a "vast majority" you could say "5 times greater" or "90%", etc.

The various maps showing emissions concentrations, depositions, etc. are very helpful. Even though effects of aerosol-phase S and N compounds are intentionally (and unfortunately in my opinion) excluded from these secondary NAAQS discussions, I think it would be useful to include maps of sulfate, nitrate and ammonium aerosol concentrations. These should be readily available as CMAQ model output, as well as from the relatively dense IMPROVE + CSN aerosol speciation networks. It would also be useful to show some "ratio" or "difference" maps – for example the ratio of S (or N) deposition to SO_2 (or NO_2) concentration and S (or N) deposition to S (or N) emissions, and the ratio of total reduced nitrogen deposition to total nitrogen deposition. Ratio or difference maps could also be a useful way of communicating differences between measured and modeled gaseous or aerosol concentrations or deposition. In making these maps, it may be necessary to employ some spatial aggregation or smoothing to "show" the information clearly.

1. The first charge question asks if the chapter appropriately identifies and describes uncertainties associated with these air quality-related analyses. The Section 3.5 "discussion of uncertainties" includes a fairly complete listing of the various types of uncertainties associated with the characterization of air quality and deposition in the case study areas. However, the descriptions and discussions of these various causes of uncertainty are so minimally detailed and (intentionally) non-quantitative that I'm not sure much useful information is conveyed.

One exception is a detailed (but graphical only) comparison of the wet N and S deposition estimates derived from simple interpolation of NADP data (the method used in this assessment) compared to similar estimates based on a higher-resolution enhanced deposition model (Grimm and Lynch, 2004), as well as to CMAQ wet deposition estimates for the Adirondack case study area and surrounding region. This is an informative comparison (and responsive to previous CASAC suggestions), but the results could be more useful if somewhat more quantitative comparisons could be included. For example, the maps showing the "similar" spatial patterns might be accompanied by scatter plots providing a more quantitative comparison of the gridded data, perhaps with different colored symbols to indicate grid points (a) within the Adirondack case study area and (b) within or including the watersheds of the case study lakes. A minor point here is that I don't think the Grimm/Lynch model really accounts for cloud water deposition, which can make substantial additional contributions above elevations of about 600m.

Section 3.3.2 indicates that the discussion of uncertainties in section 3.5 will include a comparison of (2002) results from CMAQv4.6 and v4.7, but no such comparison is actually included there. A brief mention of these comparative results is given in Appendix 1, but this is quite minimal (TNO_3 over-prediction in CMAQv4.7 is about twice that of CMAQv4.6), includes no discussion of possible causes of the differences, and is not especially useful. I recommend adding some quantitative summary (perhaps a table) of the model/measure and model/model comparisons presented in Appendix 1 to Section 3.5. This could be accompanied by a caveat that such quantitative comparisons are not intended to be comprehensive…

To the extent possible, it would be useful if some indications of relative uncertainties could be provided. For example, I would guess that uncertainties in estimated dry deposition are greater than for wet deposition, and that uncertainties in the total deposition of deposition of reduced N are greater than for oxidized N, which are greater than for sulfur deposition. In addition, I would imagine that uncertainties in the characterizations of emissions, air quality and deposition are relatively small compared to uncertainties in the resultant chemical and biological responses in the affected ecosystems.

2. The second charge question asks whether replacing the previously presented RSM analysis with a new set of CMAQ simulations provides an ability to adequately examine the contributions of NO_x emissions to total (reduced and oxidized) N deposition. I'm not sure that the previous RSM approach was inadequate; rather I think the RSM and its uncertainties were not very well explained and documented in the last draft REA. However, I do think the current approach including additional CMAQ runs, which separately explore effects of 50% reductions in NO_x, NH_x and SO_2 emissions and also explore inter-annual variability through use of different meteorological years, provides a clearer and more transparent view of the relative contributions from oxidized and reduced N emissions. This is further supported by the model/measurement and model/model comparisons summarized in Appendix 1.

3. The 3rd charge question asks if the evaluation of CMAQ model performance presented in Appendix 1 is sufficient to support the use of the model in this review. In my opinion, Appendix 1 is presented very clearly and concisely, and does provide sufficient support for the use of the model in this review. One suggestion that might help strengthen this support would be to include some additional references documenting the fairly extensive past performance evaluations to which the model has been subject (i.e. by Appel, Napelenok, Pinder, Gilliand, Dennis, etc.). A second suggestion is to add – either in the appendix or in the Chapter 3 section on uncertainties, a brief summary of the possible implications of the model performance evaluation to the risk assessments being conducted in the selected case study areas.

Specific comments on Chapter 3

p. 3-2, line 18: "$_x$" should be "NH_x". Also you could add ", and their atmospheric transformation products" after "SO_x" and add "the earth's" before "surface properties".

p. 3-2, line 19: You could change "parameters" to "variables".

p. 3-2, line 22: absence of routine occult deposition measurements is not much of an excuse for ignoring what we know very well is an important input at higher elevations and in coastal areas. You could have at least modeled it, as you have done for dry deposition.

p. 3-3, lines 3-4: This is only a half truth. NO and NO_2 are also relatively reactive and may be removed by chemical transformation (to soluble species and/or to compounds like HNO_3 with high deposition velocities) and more rapidly than less reactive more soluble gases like SO_2 under a range of conditions. Also, a high proportion of NO_x emissions are from automotive sources at ground level, and so the potential for long range transport is less than for elevated stack releases.

p. 3-4, line 11: Could you add a parenthetical (XX%) to indicate more clearly what you mean by "the vast majority".

p. 3-5, line 4: Same comment as previous one. It would cost you no space to indicate quantitatively by how much are the NO_x emissions "far greater" in the East than in the West. One easy way to do this would be to add "(total 2002 emissions = XXXX tons/yr)" to the labels of the Eastern , Western and National US NOx emission pie charts in Figure 3.2-1. Also in line 5, instead of telling us that the most of the Western NO_x emissions are from California but are "not shown", why not just add a (XX%) and show us what you mean.

p. 3-5, lines 7-8: So what's the implication (if any) for deposition-related effects of highly episodic NO_x emissions from forest fires? Do we expect episodic acidification or eutrophication? I doubt it.

p. 3-6: I don't understand how confined animal feeding operations greatly increase the <u>volumes</u> of animal wastes. I can see confined feeding increasing emissions densities and the emissions of volatile components of these wastes to the atmosphere (or in runoff to surface waters), but would think the volume of waste produced by 100 cows is about the same weather they are confined or disbursed.

p. 3-8, line 5: You could add "stationary combustion" between "non-EGU" and "sources", as everything is either an EGU or a non-EGU.

p. 3-8, line 10: How much is "much greater in the East"? Again, why not just add the East, West & National totals to the pie chart labels in Figure 3.2-4.

p. 3-10, line 13: add "." after "area".

p. 3-11, lines 27, 28, 32 and p-3-12, lines 1, 4, 7, 10: Text describing figure refers to "NO_x" but Figure 3.2-6 refers to "NO_y".

p. 3-15, lines 21-22: I assume you mean "reduced nitrogen wet deposition was calculated from measurements of wet ammonium (NH_4^+) deposition".

p. 3-16, line 13: You could add ""N" after "measured".

p. 3-24, lines 9-10: Refers to a comparison of CMAQ v4.6 and v4.7 results included in Section 3.5 discussion of uncertainties, but no such comparative results are presented there, and these are only minimally described in Appendix 1 (which includes a footnote on p 1-1 noting that the differences in v4.6 and v.4.7 are small, "as described in Chapter 3". It would be useful to actually see this description in chapter 3, along with a more detailed comparison in the Appendix.

p. 3-27 and elsewhere "Whiteface" is one word.

p. 3-27, lines 17-18: The recently available 2008 data show a decrease, and you could add a comment to this effect.

p. 3-31, Figure 3.3-3a: I think it would be clearer and shorter to present this data and that from the following 8 pie charts in tabular form and to show both the amounts in kg/ha/yr as well as in %, or you could at least add the totals (X kg/ha/yr) in the figure titles.

pp. 3-49 through 3-57: I would prefer that figures 3.3-6a through 3.3-6i were stacked bar charts showing the absolute (rather than %) amounts of the various components (Wet Re N, dry Ox N, etc.) in each season. We don't need the units to be "% of annual total by season" to see which seasons are most and least important and by how much. Similarly, the charts in figures 3.3-7a through 3.3-7i don't seem to convey very useful information. I think deposition of some species may have different environmental consequences in different seasons, and so it would be more important to no how much of spring N deposition comes from oxidized N than it is to know what fraction of total annual oxidized N is deposited in spring. Also, if absolute amounts were shown rather than percentages of total, it would give a good sense of differences in the various kinds of deposition by site and by season.

p. 3-59, line 10: Whiteface is one word.

p. 3-62: Use consistent terms for the case study areas in the 2 figures (Transverse Range vs. Los Angeles Range).

pp. 3-70 through 3-78: As for the seasonal N deposition charts, I think stacked bar charts showing absolute amounts from wet & dry S dep. by season would convey more useful information, and would also convey the differences in total & seasonal dep. rates in the different study areas. Also, what's the purpose of the large gap between dry & wet dep. bars in figures 3.3-15a through i?

p. 3-81, line 15: It isn't clear what "This" refers to.

p. 3-82, line 5: "$_4NO_3$" should be "NH_4NO_3".

p. 3-82, line 8: "$_x$" should be "NH_x".

p. 3-82, lines 14 & 15: Figure 3.4-4 doesn't really show "contributions of total reactive nitrogen approaching 50%". It shows a nearly 50% reduction in total nitrogen deposition if NH_3 emissions were reduced by 50%. If contributions of 50% were reduced by 50%, total N dep would have been reduced by 25%.

p. 3-82, line 29: "$_4NO_3$" should be "NH_4NO_3".

p. 3-93, line 5: I think "similarities in differences" should be "similarities and differences".

pp. 3-94 – 3-96: The figure captions refer to "total" oxidized N, reduced N or sulfur deposition, but I think they are (and text indicates they are) for "wet" deposition only.

Specific comments on Chapter 4

p. 4-5, line 17: I think you mean "…models do a better job projecting ANC than they do for projecting pH and inorganic Al …"

p. 4-1, Figure 4.2-4: The font labeling the figure axes and legend is too small to read.

p. 4-14, Table 4.2-2: Give units in table (μeq/l, I presume).

p. 4-18, line 7: Table 4.2-2 does seem to indicate that current SO_4^{2-} is "19 fold" greater than NO_3^- but this seems inconsistent with Figure 4.2-6 – which shows current (2006) SO_4^{2-} is about 4 times greater than NO_3^-. Why the difference?

p. 4-24, line 1: Should be "reductions … are" or "reduction …is".

p. 4-25, lines 24 & 25: Table 4.2-4 needs units, and again the observation that SO_4^{2-} concentration is 11 fold greater than NO_3^- (from table) is inconsistent with figure 4.2-14 which shows current (2005) sulfate being about 6x nitrate.

p. 4-40: It would be more effective to show some quantitative results from the sensitivity analysis in tabular or graphical form – possibly in an appendix – rather than just claiming that you did it and that it looked good to you. Without better context and detail, its impossible to know the implications of some of your summary statements – for example what does it mean that "similar results were given for the number of lakes with all realizations above the critical load" or that "changes in critical load values could range from 3 to 24 meq/m2/yr…"

p. 4-41, lines 13-18: Its not clear what's being described "which is on average a 15 μeq/l difference in ANC concentrations or 10%" or what's "on average [an] 8 μeq/l difference in ANC concentrations or 5%".

General comments on Chapter 7
Synthesis and integration of these disparate case study results is an inherently challenging undertaking, and given the difficult task, I think chapter 7 is reasonably well done. I especially like the discussion of uncertainties in Section 7.2 which is organized along the lines of a conceptual model which ultimately relates ecological benefits or welfare effects back to ambient air quality indicators. This conveys the logical progression from air quality to environmental effects and also illuminates the weakest links in this chain. Since the case studies are both diverse and also represent rather extreme conditions, its difficult to envision how the case study results might be scaled to cover a range of environmental effects extending over broader spatial areas. For aquatic acidification, examples are provided for how the case study results might be extended to larger populations of lakes and streams, but its much less clear how this could be done – in ways that would either support selection of specific air quality indicators, or support implementation of secondary NAAQS based on those indicators – for terrestrial acidification or aquatic or terrestrial nutrient enrichment. Some added discussion of the spatial extent of the kinds of environmental effects considered in the case studies would be useful in this chapter.

Another possible missing piece in this synthesis and integration is a discussion of how the different environmental indicators might relate to each other. Using the example conceptual model diagram in Figure 7-2, it seems like there might be (at least) four different diagrams and secondary standards - one each for aquatic and terrestrial acidification and aquatic and terrestrial nutrient enrichment. And each of these would be further modulated by various mixes of S and N deposition and the inherent sensitivities of the effected ecosystems. However, it also seems likely that there would be substantial overlap in areas with surface waters exceeding an ANC threshold and areas where forest soils have Bc/Al ratios below a specific threshold. Possibly the two indicators are sufficiently closely related that a single indicator could be selected that would be "controlling". I wonder if it would be possible to identify a Bc/Al ratio that's the approximate "equivalent" (i.e. would cover about the same spatial area) of an ANC of 50? Or in terms of the severity of environmental effects or the sustainability of some current or improved level of environmental quality, what level of ANC is the approximate equivalent of a Bc/Al ratio of 10, and what kinds of ecological effects from aquatic or terrestrial N enrichment would be considered to be of similar severity? For a given "acid sensitive" area with poor buffering capacity, it would be useful to know whether the aquatic acidification threshold or the terrestrial threshold is more limiting (and whether the answer to this question is based on the inherent environmental sensitivities or on the relative severity of the selected thresholds of adversity).

1. The first charge question asks about the extent to which the description of ecosystem services provides a useful framework in the case study analysis for informing standard setting. While I think the quantitative estimates of (selected) ecosystem services can be one useful consideration to inform standard setting, I think it should be clearly recognized that each presented example of ecosystem services is itself a limited case study. I think the example of considering (only) sport fishing (dis-) benefits from aquatic acidification helps illustrate this point – as it seems only a few degrees removed from David Stockman's infamous 1983 calculation that acid rain controls would cost $6,000 per pound of Adirondack brook trout. I think the presentation of example ecosystem services is useful in chapter 7, but it should be emphasized that only a few examples – with relatively easily monetized benefits - are provided here. An alternative approach to considering "adversity" of effects might be to consider the concept of "sustainability". Given geo-specific rates of soil & bedrock weathering for example, what rates of acidic deposition can be sustained indefinitely without eventually leading to degradation of environmental resources. I think this concept of sustainability is especially important in considering effects which result from long-term, cumulative deposition processes.

Another relevant concept which is not really addressed in this chapter or in previous sections relates to the nature and/or rates of "recovery" of adversely affected ecosystems. A current level of damage that has resulted from past and continuing deposition of acidifying or nutrient enriching S and N compounds may not be fully reversible to pre-deposition conditions and/or may require a substantial lag period between reduction in emissions (and concentrations and deposition) and improvements in environmental conditions. Possibly the "time to" and "degree of" recovery might be variables considered in judging the relative adversity of effects. Since secondary standards have no fixed time requirement for attainment, another possible approach here might be to consider standards requiring a certain rate of progress towards long-term goals which may not be feasible to attain quickly in all areas.

2. The second charge question asks if sufficient information is provided to inform standard setting based on effects other than aquatic acidification (terrestrial acidification, terrestrial nutrient enrichment and/or aquatic nutrient enrichment). As indicated above, I think its possible to identify some deposition thresholds estimates of terrestrial acidification effects, using something like the Bc/Al ratio, that might be extended over relatively large spatial areas (as has been recently done for New England and Eastern Canada and as is currently being extended to NY). I also think its likely that there will be considerable overlap in regions of, and extent of severity of effects on, aquatic and terrestrial acidification, and/or that there may be a common level of S+N deposition that provides an approximately equivalent level of protection against both kinds of effects. If a specific ANC threshold is considered for aquatic effects, it should also be relatively easy to calculate necessary S+N deposition reductions to attain such thresholds and then to calculate improvements in Bc/Al ratios and terrestrial effects that would result.

Another possibility that should be considered is that if the ambient air quality indicators are required to be in units of the nominal gaseous criteria pollutants – SO_2 and NO_2 – such measures are more related (spatially and temporally) to the emissions that need to be controlled, while the effects of those emissions are more related to the long-term deposition of the chemical transformation products – $SO_4^{=}$ and NO_3^- – in combination with inherently sensitive bedrock and soils. These areas of greatest effects are therefore inevitably displaced, sometimes at considerable distance, downwind of the areas of highest emissions and gaseous precursor concentrations. An exceedance of an environmental indicator should therefore logically trigger exceedances of the air quality indicator in relatively large upwind regions. If for example an ANC or Bc/Al threshold is exceeded in the southern Green Mountains of VT – as can be expected given recent TMDL calculations for acid sensitive lakes there, and recent Bc/Al calculations by the NEGECP for that area – a zero-out of all SO_x and NO_x emissions in VT would have little or no effect on reducing the S + N related exceedances of the ANC or Bc/AL thresholds in that region (which is much more dependent on emissions from NY PA and OH). The secondary SO_2 and NO_2 standards should be considered to be exceeded until they are reduced to such levels throughout the eastern US that the deposition-related indicators are not exceeded in (VT and other) acid sensitive areas within that region.

Regarding possible NAAQS based on nutrient enrichment indicators, the various terrestrial effects at different and widely varying N deposition rates as summarized in Figure 7.1-3 provides some basis for considering secondary NAAQS based on several different (low, medium and high) degrees of protection based on total N deposition rates, and calculations could be made of the necessary reductions in NO_2 concentrations and NO_x emissions over broad spatial scales needed to attain those levels of protection. As with acidification-based standards, I don't think its necessary – and may be counter-productive – to try to link environmental indicators and air quality indicators on a point-specific basis, but rather we might consider what levels of total N deposition and effects protection would be achieved in areas with the most sensitive ecosystems if ambient NO_2 concentrations (and NO_x emissions) were reduced by various amounts or percentages throughout the surrounding region.

Considering a secondary NO_x standard based on aquatic nutrient enrichment is further complicated by the large and often dominant contributions from non-air sources. However, calculations could still be made of improvements in aquatic effects that would be expected if oxidized N deposition (and/or NO_2 concentrations) were reduced to certain absolute levels or by certain percentages, with contributions from NH_x and non-air sources taken as a given. This process along with similar assessments for terrestrial nutrient effects and acidifying effects of N deposition might lead to considering a range of secondary standards that would yield a range of beneficial environmental effects.

I think the discussion of uncertainties in section 7.2, organized as a conceptual model linking air quality indicators to ecological effects, can also be used as a guide to areas that would benefit most from future research and/or environmental assessment for future NAAQS revisions. I also think continuation of the work conducted for this assessment has good potential applications in areas outside the specific SO_x and NO_x NAAQS revision process. Historically, major reductions in SO_x and NO_x revisions have resulted largely through national and regional programs justified by benefits in a wide range of health and environmental effects. This will likely hold true in the future as future revisions to national automotive emissions standards, (whatever replaces) the Clean Air Interstate Rule, future checkpoints for the Regional Haze Rule, and likely lower NAAQS for PM and ozone will all result in, or be strengthened by advance assessments of, the environmental benefits of lower SO_x and NO_x emissions.

Specific comments on Chapter 7

p. 7-1, lines 21-22**:** I don't think you really mean that a welfare effect is what "society views as beneficial". Society views good visibility or a healthy environment as beneficial. A welfare effect occurs when these beneficial welfare or environmental goods are adversely affected.

p. 7-10, lines 1-7: Its not really clear if or how you intend a single ANC indicator threshold to be protective against seasonal or episodic variations. Nor does it seem likely that a given ANC limit, measured at summer low flow for example, would protect all lakes and streams equally against episodic acidification effects from spring snowmelts.

p. 7-11, lines 16-19: For the Bc/Al ratios of 0.6 and 1.2, quantitative estimates of reduced growth are provided (and seem rather "non-protective" if they would allow 75% or 50% of North American tree species to experience growth reductions). By contrast, not much justification is provided for the selection of the more protective Bc/Al ratio of 10 – other than the observation that it's the most conservative value used in some analyses. There isn't really any better justification for the 10 ratio provided in Chapter 4 or in Appendix 5, and I think there needs to be a clearer explanation for why a number of prominent studies have used this ratio. This seems especially important given the large range between 1.2 and 10, and the large spatial areas that will presumably fall within those extremes.

p. 7-12, line 4 "res" should be "red".

p. 7-13, Figure 7.1-2: This figure is not described very well in the text. For example, what are the OEC and OHI scores, or how does the chart show that a greater than 100% reduction in atmospheric N deposition was needed to move from bad to poor.

p. 7-14, line 22: What's a "compared forest health decline"?

p. 7-14, line 26: Its not clear what "(generally at deposition rates of < 10 kg N/ha/yr)" means here. I assume this is a general description of the N additions applied in a particular set of experiments – that have shown transient growth increases followed by increased mortality at deposition rates below 10 kg N/ha/yr, but don't mean to imply that experimental treatments at 10 kg N/ha/yr or greater showed no transient growth increases or increased mortality.

p. 7-19, line 12: I assume that non-use (existence) services would also be affected by aquatic or terrestrial acidification or terrestrial nutrient enrichment.

Mr. Dave Shaw

Executive Summary

1. ***In response to the Panel's review of the first draft Risk and Exposure Assessment, we have included an executive summary of this document. Does the Executive Summary adequately summarize and characterize the key issues driving this review as well as the important findings of the analyses? Does the Panel have any suggestions for clarification or refinement of the Executive Summary?***

I feel that the REA Executive Summary provides a clear summary of each of the chapters with conclusions allowing the reader to extract highlights of the report without reading the entire report.

I would like to see some clarification on critical loads and how it is used in this assessment. I appreciate that the areas of confidence are expressed in the conclusions.

General Comment

I am still committed to the idea that one of the key issues driving this review should be to also identify where data is missing with the intention of filling those gaps. I feel that this should be clearly stated in the Executive Summary, perhaps also listed in the Policy Relevant Questions on Page ES-3. While this may be somewhat addressed in the last two bullets, I feel that it deserves a little more emphasis.

Some other items that would help to emphasize this key issue:

- Improve the link/reduce uncertainties between ambient air quality indicator and deposition.
- Improve/reduce uncertainty in response models (e.g. MAGIC and SMB) or create new ones, to be better predictors (e.g. of pH, Al) and to be better at scaling up to the broader regions. Consider developing Base Cation Surplus as a substitute for ANC.
- The above bullets would require improved input data.

The natural course of action in addressing the above bulleted issues would be to ask EPA to make a separate exercise of identifying future data refining needs. Now that we have the ISA and REA documents to substantiate the needs. The goal should be to increase the confidence in ecological exposure, responses, effects and benefits *to all other areas beyond aquatic acidification*.

I propose that EPA sponsor or provide for a focus group on monitoring and research information that would reduce uncertainty (by improving the input data and improving the models themselves) from these case study areas for national assessments of CL. Uncertainty analysis of those results should be ongoing in the CL process.

Chapter 3: Sources, Ambient Concentrations, and Deposition

1. ***This chapter describes an approach for characterizing the spatial and temporal patterns of nitrogen and sulfur deposition in the case study locations including both oxidized and reduced nitrogen, and both wet and dry deposition of oxidized nitrogen and sulfur. Are the uncertainties associated with these analyses appropriately identified and described?***

The uncertainties associated with these analyses are appropriately identified and described, with a few possible exceptions. In terms of air concentrations, was there an attempt to compare modeled NOx and SOx where measurements are available? Even though there are few NO_2 monitors, and most are in urban areas, it would help to see that at least in urban locations, the model predictions of NO_2 were reasonable. There are still numerous SO_2 monitors at SLAMS/NAMS sites as well.

Section 3.5 does mention some of the issues related to combining measured wet deposition with modeled dry deposition, but leaves out at least one limitation. The spatial analysis using NADP wet deposition data does not capture urban influences well (if at all), whereas the modeled dry deposition will include urban sources and influences (that, of course, are not measured routinely). This methodology may overemphasize dry deposition in the vicinity of large point sources or urban/suburban areas, and we don't have adequate data to assess how important this is.

2. ***In response to CASAC's recommendation, the RSM analysis presented in the first draft Risk and Exposure Assessment was replaced by an analysis of results from a new series of CMAQ simulations designed to explore the relative contributions of NO_x and NH_3 emissions to total, reduced and oxidized nitrogen deposition and the relative contribution of SO_2 emissions to sulfur deposition. Does this approach enable us to adequately examine the contribution of NO_x to total nitrogen deposition?***

Yes, the 50% NOx and 50% NH_3 reduction scenarios are generally adequate to quantify the contributions of NOx to total N deposition across the modeling domain. However, Section 3.4 has a disconnect with the rest of this chapter of the draft REA. In this section the wet deposition is taken directly from the model, whereas in previous sections the wet deposition is derived from the NADP data. This is necessary in this section since you are trying to estimate changes in modeled wet and dry deposition resulting from changes in modeled emissions.

3. ***The CMAQ application and model performance evaluation is presented in Appendix 1, as recommended by the Panel. Is this analysis sufficient to support the use of the model in this review?***

The analysis presented in Appendix 1 is generally sufficient to support the use of CMAQ in the draft REA, especially with regard to wet deposition. The seasonal, year-to-year, and broad geographic variations are clearly displayed. On the "dry" side, the model evaluation suggests that ambient concentrations are reasonably well characterized. However, dry deposition is a significant contributor to the total loading of S or N, and it is not clear that the CMAQ dry deposition estimates are valid or even reasonable. While the CASTNet program does not measure dry deposition directly, it still might be useful to compare model predictions of dry deposition at CASTNet sites.

Chapter 4: Acidification

1. ***Section 4.2 and Appendix 4 describe the analyses used to evaluate the effect of aquatic acidification. The analysis evaluates the ANC in selected lakes and streams in the Adirondacks and Shenandoahs relative to three potential ANC cutoff levels (20, 50, and 100 µeq/L) to determine the impact of current levels of deposition in these areas as well as a larger assessment area. Is this data adequate to establish critical loads of deposition for the case study area?***

ANC is a good indicator and these are good cut off levels for selected lakes in the Adirondacks and selected streams in the Shenandoahs. However, these lake standards may not be protective of streams in the Adirondacks. For instance, we know from ongoing research that Adirondack streams do not respond the same as lakes and are not experiencing the recovery that has been detected in lake trends. We are also learning that Base Cation Surplus is a better indicator than ANC to characterize biological impacts in streams. This was mentioned in the ISA (Lawrence et al) and should be reported in the REA.

There are aspects of the Adirondack Case Study Area assessment that need clarification including:

1. Respresentativeness of the modeled waters
2. The treatment of natural acidity
3. Review of the paleolimnological data on preacidification conditions

Representativeness of the modeled waters

The modeling discussion is not clear about the various Adirondack data sets used. There is not enough explanation as to which lakes were used from what survey program and why. For instance (pg 4-14) MAGIC was run on 44 lakes in the Adirondack Case Study, said to be taken from the EPA TIME (n =43 lakes) and LTM (n= 52 lakes). It would be helpful to explain why some lakes were chosen and not others.

It appears that average yearly ANC values were calculated from single measurements, because Adirondack TIME waters are collected only once per year. This should be explained more clearly.

Page 4-16, shows 169 lakes were modeled in the Adirondack Case Study Area. Figure 4.2-9 (pg 4-20) shows ANC values from 94 TIME/LTM lakes, however there are only a maximum of 89 TIME and LTM lakes due to the 6 overlap waters between the two.

The treatment of natural sources of acidity in lakes

Natural acidity is a common phenomenon for the Adirondack Lakes region, this, combined with the observation that DOC is increasing in some areas, demonstrates that more consideration is required than what is provided here. I am unclear as to how a 'natural' ANC concentration of less than 50 determined (pg 4-34 line 26), perhaps more explanation is in order.

Review of the paleolimnological data on preacidification conditions
Were any of the selected MAGIC lakes, part of PIRLA (Paleolimnological Investigation of Recent Lake Acidification) lakes where diatom sediments provide for reconstructed histories of lake chemistry to prior to the onset of recent acidification?

2. ***The ecological effect function for aquatic acidification (section 4.2.7) attempts to characterize the relationship between deposition and ANC. In order to estimate the amount of NO_x and SO_x deposition that will maintain an ANC level above a given limit requires the knowledge of the average catchment flux of base cation from weathering of soils and bedrock (i.e. preindustrial cation flux (BC_0)). How might we generalize from location specific inputs (F-factor approach) to using this approach on a broader scale – watershed, regionally, or some other way – to generalize beyond individual locations? What other methods should be examined for estimating catchment weathering rates nationwide for surface acidity?***

The link between deposition and an ANC in surface water is crucial and difficult not only at each specific location but also in order to generalize findings on a broader scale. Determining catchment weathering rates is critical because 'the catchment supply of base cations from the weathering of bedrock and soils is the factor that has the most influence on the critical load calculation and has the largest uncertainty" (pg 4-40 lines 3-5) contributing almost half of the total variability in critical load estimates (pg 4-69 line 16).

As far as methods to examine this and the generalization from specific inputs/sites to apply to a broader scale, I note that current research is underway through the New York State Energy Research and Development Authority by Miller, Lawrence, Weathers and others to examine weathering rates and these regionalization questions. These results and other related work e.g. sponsored by the National Park Service should be incorporated as they are made available

Aquatic acidification is the best place to start because of the weight of the evidence across many sensitive areas. However, while ANC is currently the indicator of choice, other more biologically direct indicators (pH and Aluminum) should be considered along with the newly developed Base Cation Surplus. These will likely be more useful further down the road in the critical loads process. For example, inorganic monomeric aluminum at detectable levels (2 umol/L) is toxic to biota in any environment, aquatic or terrestrial. Its presence is directly associated with anthropogenic acidic deposition.

3. ***Section 4.3 and Appendix 5 describe the analyses used to evaluate the effect of terrestrial acidification. This analysis uses the Simple Mass Balance Model to determine the impact of current deposition levels on Bc/Al levels relative to three potential Bc/Al cutoff levels (0.6, 1.2, and 10.0) for sugar maple in the Kane Experimental Forest and red spruce in the Hubbard Brook Experimental Forst and a larger assessment area based on the FIA database for 17 states. Is this approach adequate to develop critical loads of deposition for the broader terrestrial acidification case study area? Is the regression analysis between Bc/Al ratios and tree health sufficiently described and are uncertainties adequately characterized?***

The approach is adequate to develop critical loads for these case study areas, but I feel that further discussion is needed to determine whether the assessment is adequate to inform about the terrestrial responses of all sensitive areas like the Adirondacks and other US regions (Fig 4.3-3) with potential red spruce and sugar maple sensitivities. It would be helpful to explain how the indicator soil solution ratios at the Kane Experimental Forest (PA) and/or the Hubbard Brook Experimental Forest (HBEF) in NH are going to be translated to protection of sensitive sugar maple and red spruce ecosystems in other regions? Specifically, how will the uncertainties with misclassification of soil parent material (pg 4-69) be resolved?

While HBEF is a pre-eminent long term forest ecosystem study site, it has limitations representing high elevation northeastern forests. For example, the high elevation forests (greater than 900 m above mean sea level) of the Adirondacks cover over 100,000 acres and according to Miller et al 1993 have accumulated 80% more sulfur than the lower elevation forest at HBEF. A significant portion of the sulfur as measured at Whiteface Mountain in the Adirondacks comes from dry deposition and cloudwater contributions.

A question that may be in order for discussion is: Are the number, location and routine parameters collected by the USFA FIA permanent sampling plots adequate to support the analysis for acidity critical loads to high and medium elevation forests in the northeastern US?

Chapter 5: Nutrient Enrichment

1. ***Section 5.2 and Appendix 6, describe the analyses used to evaluate the effect of aquatic nutrient enrichment. The analysis uses the SPARROW model on one stream reach (Potomac River and Neuse River) to determine the impact of atmospheric total nitrogen deposition on the eutrophication index for the estuary. Does the Panel think that the model is adequately described and appropriately applied?***

I feel as if SPARROW is adequately described within the REA and appreciate the detail in Appendix 6. I especially appreciate the use of the New England waters paper by Moore et al.

I believe it would be helpful if in either the REA or Appendix 6 a table with the data type and sources were printed. For example, where was the loads data derived from, or the stream network? The use of the other tables in this section are especially helpful, so this additional table would contribute to the ease of understanding.

Case Study Analyses (Chapters 4 & 5)

2. ***In using the Risk and Exposure Assessment to inform the policy assessment, we plan to focus on aquatic acidification as the basis for an alternative multi-pollutant secondary standard as this is the area where we have the most confidence in our ability to characterize adverse effects. Does the Panel agree with this approach?***

I feel that aquatic acidification is the best place to start because of the weight of the evidence across many sensitive areas. However, while ANC is currently the indicator of choice, I believe that other more biologically direct indicators (pH and Aluminum) should be considered along with the newly developed Base Cation Surplus. These will likely be more useful further down the road in the critical loads process. For example, inorganic monomeric aluminum at detectable levels (2 umol/L) is toxic to biota in any environment, aquatic or terrestrial. Its presence is directly associated with anthropogenic acidic deposition.

Specific comments/questions

Chapter 3: There are several instances in which "NH" is left out – Page 3-2 lines 9 and 19, and Page 3-82 line 5.

page 4-10 Figure 4.2-2 New England is not an ecosystem

Pg 4-15 Figure (a) How were the critical load (green dot) modeling sites selected?

Pg 4-14 line 14. What is the present condition? Here it is defined as 'present (2002 and 2008)' whereas the discussion starting on pg 4-18, calls 2006 the current condition. Which are actual measured and which are modeled current condition values?

Pg 4-39 Uncertainty and variability. How well do the MAGIC model simulations for current ANC concentrations measure against measured median ANC?

Pg 7-1 lines 21 to 27. By transitioning to the "ecosystem services" concept, (we must also preserve) there is a danger of losing the broader definition of effects that are important to people or that society views as beneficial. Specifically it should be recognized that the majority of the sensitive areas identified in these documents are either national or state parks or scientific experimental forests that they have a collective intrinsic value that have been previously identified as exemplary. They deserve the bar of level of protection to be raised. (Protecting biota in a park or a scientific reserve is 'more important' than anywhere else because we have as a society identified these are part of the public good.)

Pg 7-10 lines 5 to 7. Crucial point but sentences are awkward. Not exposure to an annual mean, but on average with an annual mean of 50, the probability is low that the lake will go down to 0 during springmelt. The length of time sentence should be deleted as it does not summarize that section, but a point to be made as part of the background to this discussion.

Appendix 1, Footnote 8: Why is NOy listed as the sum of NO, NO_2, HNO_3, and PAN? It should include also HONO, N_2O_5, ANO_3, and NTR as is listed in the Table 1.1-2 for deposition.

Appendix 1, Section 2.1 and Figures 2.1-1 through 2.2-7: Don't these additional lines indicate a factor of 1.5 around the 1:1 line (not a factor of 2)?

Discuss further the consequences of MAGIC using a calculated ANC rather than measured Gran ANC. How well does MAGIC scale up beyond the watersheds of the modeled lakes themselves?

Dr. Kathleen Weathers

I appreciate the enormous work involved in putting this REA together. It was (and is) a Herculean effort.

I have included brief comments—some overarching, some specific—on many of the chapters in the REA, and on the charge questions.

Executive Summary

While I like the idea of Figure ES-1, I think some of the pieces are a bit misplaced and mislabeled. For example, mobile anions might be depicted in association with cation leaching, and to what are "soil processes" meant to refer (e.g., NO3—meaning nitrification?).

Figure ES-2. I am a fan of using multiple colors in Figures only when their purpose(s) are clear, or clearly identified. At first glance, I'm not sure what the different colors are meant to depict (if anything) in this, and other figures in this document. Ditto for shapes. I suggest either identifying the significance of both in the Figure legend, or (re)considering carefully their use.

Figure ES-4: The recreational fishing example here, and throughout the document, in fact, would benefit from a footnote, or a few lines in the text that give a clear explanation about why and/or how the effect of fish stocking or invasions, an important driver in species distributions and abundance, can be separated from the effects of acidification via deposition.

Pages ES-12, 13—it would be a good place to note some brief caveats about where this spatial estimation is likely to underestimate (or perhaps overestimate) deposition (e.g., high elevation ecosystems, heterogeneous terrain, and regions with significant snowfall). It would also be useful to note in the figure legends what the spatial resolution is for these maps.

Page ES-16 (line 14): I think that MAGIC and other models are used to estimate (vs determine).

Note how the "critical load approach" boxed here either parallels, or does not, that used by Europeans and others.

I think it very important to note, and put in context, when, where, and how natural (vs anthropogenic, or strong mineral) acidification occurs (terrestrial and aquatic). (The language exists in chapter 4.) Nothing more than a brief mention is necessary in this Executive Summary.

What determines when references are included, or not included, throughout the REA and especially appendices? Mostly the text is not referenced (beyond the ISA), except when it is!

Chapter 1:

There is reference to "the latest scientific knowledge," yet many of the references I noticed in this document are from quite some time ago.

1-10, line 26: the term “total nitrogen” should be qualified here. Sometimes total is referred to as having captured all the vectors of N, i.e., having estimated dry, wet and cloud or fog deposition.

1-12, lines 18-19??

1-14, lines 4-6 and a few other places in this document: In these systems, atmospheric N deposition is the main source of new N to the system, but it may also travel through vegetation and soil before getting to headwater streams and lakes, depending upon where they sit in the landscape and by what they are surrounded. Perhaps what was meant here was headwater streams and high elevation lakes that are above treeline? This statement should be clarified here and elsewhere in the document.

1-19: I agree with the paragraph starting on line 7 (and the bulleted list preceding it), and thus Figure ES-4 (depicting relationships) challenges my sensibilities. I’ll be interested to be reminded of its heuristic utility.

Chapter 3:

I suggest adding the spatial scale/resolution to the figure legends of all of the figures that show spatial data.

3-2: While it is reasonable that cloud or fog deposition could not be considered for lack of monitoring data, there are places throughout the document where the qualitative result of this (necessary) omission could be pointed out (e.g., for example, eastern high elevation sites where cloud/fog has been shown to contribute significantly to N and S loads).

3-24. It would be useful to report under “spatial allocation of gridded data to case study areas” how many (roughly) grid cells were used/study area. Given the coarse resolution of the CMAQ data, it seems possible that the Hubbard Brook Experimental Forest, for example, could contain few grid cells.

3-26: For the data that span a 4-year time period, “trend” analysis is dubious. Analysis of the long-term record at Hubbard Brook suggests that detection of statistical trends took more than 3X that time period.

3-90s: I expected to read more about the uncertainties associated with complex terrain in this section. It is one of the important drivers in uncertainties for estimating dry deposition and a major challenge to estimating deposition.

Chapter 4:

Overall, I found most sections of this chapter to be confusing, and in need of significant editing--for clarity, for completeness, for accuracy, especially in the aquatic section. As a result, I have not made detailed comments, rather I have identified some general (and a few specific) issues.

It might be helpful to discuss charge balance early on.

As I've suggested before, defining what is meant by such terms as "health" of terrestrial and aquatic ecosystems is important if the word is going to be used.

I suggest adding a section on terrestrial-aquatic linkages.

There are many figures in this section (and in the Appendix) that are identified as showing declines (by x%, for example) over time, yet no regression, or trend analysis seems to have been performed, rather a line graph has been generated and trends/declines suggested. In the case of the associated appendix, lines may have been fit to the data, but there is not information about what kind of a polynomial fit may have been used, why, and what its significance is, for example. It is important to support the assertion of trends or declines over time with appropriate trend analyses and documentation.

I like the idea of using boxes. However, many of the boxes need editing as well.
The box on page 4-16 was particularly convoluted.

4-15: critical load approach –there seem to be a few different definitions and interpretations throughout the REA. It would be helpful to standardize.

4-17: I assume signs of improvement mean that NO3 and SO4 have decreased in surface waters of the ADK case study. See my comments, above, about the lack of trend analyses.

4-24: what are "slight" signs of improvement, or what does it mean to have "not resulted in much improvement?"

Figure 4.2-21. Fix spelling on title.

Terrestrial Effects

4-36: what's it mean to account "for the effects input of Cl-?

The connections between tree "health," direct and indirect tree responses, and growth responses should be made clear. And again, it is necessary to define what is meant by health (box on 4-44).

4-45. Many things affect the provisioning of services, of course. Some contextual statistics would be useful in this section, especially about some of the other factors that might affect these two species' productivity.

4-51: What percentage of the area used for the HBEF is covered by red spruce?

4-61: what area (ballpark) do the FIA plots cover for each of the species?

Chapter 7, Synthesis:

Much of the first part of the synthesis chapter is a reiteration of other chapters in the REA. I think that the focus should be more actual synthesis among these chapters.

I prefer the use of ecosystem structure or function (vs processes, as used throughout the REA).

Is change the right operative word/focus here?
7-2, line 13 changes can be or are quantified? tense?

Figure 7-1: see comments above about the use of color, or not. In this figure, color obscures the text.

7-3, line 19, Knowledge about the relationships, whether direct or indirect,...
Foliar injury is not really a process. Perhaps it's the change in structure that affects a process (photosynthesis) that is the point of this example? The examples at the bottom of this page could be clarified.

7-5, first para: I like the idea of this—clarifying which data are best suited to informing a policy on welfare effects, but it wasn't clear to me the current answers, i.e., I was looking for a clear path to answering the question.

7-5: Nitrogen is known to limit growth and productivity. S is necessary, but not identified as limiting, usually. The text should be changed. Reproduction of ecosystems? *Anthropogenic* acidification *can* cause...This paragraph could use some editing.

See earlier comment on the statement that atmospheric nitrogen deposition is the main source of new N. It's the main source of new N to ecosystems, and subsequently to most headwater streams...

7-6, line 1: Nitrogen deposition can alter ecosystem processes, such as primary production and nutrient cycles...

Figure 7.1.3 needs a little more description in the legend. Are the deposition ranges from the modeling and estimates in this REA, or are they estimates provided in the studies cited for the effects noted? I like the idea of this kind of figure that can be used to synthesize across systems and depositional loads.

7-17, line 6, will remain unmonetized because?

In regard to some of the specific charge questions asked:

Case studies:

Uncertainties: I am inclined to ask what kinds of uncertainty analysis are necessary from the policy making perspective, as well as how best the myriad scientific uncertainties that exist can be communicated in such a way that it is obvious which are the most "important" (e.g. have the biggest effects). Finally, I would ask how uncertainties can be communicated in such a way that they do not undermine what is known. Currently, I do not think that uncertainties are as well characterized as they might be.

I agree that it is in aquatic systems where the most direct and clearest effects of acidifying deposition have been shown. However, Chapter 4 should be significantly improved to make it useful to that process.

Acidification:

The first question is tied to the definition of critical loads, and critical loads approach. Isn't the question really are these the critical loads?

Synthesis:

The idea of ecosystem services focuses on the work that ecosystems do in service of humans. A focus on ecosystem services can limit understanding of ecosystem structure and function. However, it has purchase in the scientific community. And, in the REA the definition of welfare effects given on page 7-1 underscores the fact that discussing ecosystem services may be a useful way for scientists to translate ecological function into welfare effects.

Understanding will be enhanced significantly through generating better estimates of (all important chemical forms of N and S, especially NHx and organic N) deposition (including wet, dry and fog) to complex ecosystems, through long term experimental manipulations, and process studies that included linked biogeochemical studies.