



UNITED STATES ENVIRONMENTAL PROTECTION AGENCY
WASHINGTON D.C. 20460

OFFICE OF THE ADMINISTRATOR
SCIENCE ADVISORY BOARD

May 19, 2008

EPA-CASAC-08-012

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

Subject: Clean Air Scientific Advisory Committee's (CASAC) Peer Review of EPA's *Integrated Science Assessment (ISA) for Oxides of Nitrogen and Sulfur – Environmental Criteria* (First External Review Draft, December 2007)

Dear Administrator Johnson:

The Clean Air Scientific Advisory Committee (CASAC) NO_x & SO_x Secondary National Ambient Air Quality Standards (NAAQS) Review Panel met on April 2-3, 2008 to review EPA's *Integrated Science Assessment (ISA) for Oxides of Nitrogen and Sulfur – Environmental Criteria* (First External Review Draft, December 2007) (see Appendix A for the Panel roster). This letter has been reviewed and approved by the chartered CASAC at the public conference call on May 5, 2008 (see Appendix B for the CASAC Roster). While the Panel views that the first draft ISA was informative, it also found that the first draft did not fully meet its objective. The ISA should be an informative, succinct, and useful summary of the evidence for consideration in the NAAQS review process. The Panel recognizes that this is a significant undertaking, particularly noting that this is the first effort to consider developing standards that might integrate across multiple criteria pollutants as has been recommended by a number of advisory bodies, including National Academy committees. In this letter, the CASAC Panel first offers general comments and recommendations followed by responses to the Agency's charge questions. Comments from the individual Panel members are provided in Appendix C.

1. An Executive Summary would be valuable. It should bring forward the key points and findings from the individual chapters, with emphasis on the summary and conclusions. In addition to highlighting the findings and "take home" points from the ISA, an Executive Summary would also provide various linkages between the pollutants under consideration, their atmospheric dynamics, and their environmental effects. Further, the Panel recommends that the introductory chapter also include a table and associated discussion of which effects are or are not being addressed in this NAAQS review and

why. This table and discussion should also identify where the associated information can be found in the ISA and in what other documents the effects that are not included are discussed (such as the PM ISA). Additionally, a summary of the data and studies that would reasonably be used to inform the setting of the secondary NAAQS should be presented in a second table (or tables). The Panel recommends that the critical data needs be included both here and in the Conclusions chapter.

2. An issue pertinent to all of the chapters is an imbalance in how the various species of total reactive nitrogen¹ (Nr) (which includes oxidized, reduced and organic forms of nitrogen) are treated. At present, there is significantly more emphasis and information for oxides of nitrogen than for reduced and organic nitrogen compounds. Greater consideration should be given to the emissions, dynamics, effects and linkages of reduced and organic nitrogen based on the current and growing recognition of the importance of these forms of Nr in the biogeochemical N cycle with respect to nitrogen enrichment and acidification.
3. The effects of Nr should also be considered in the context of multi-pollutant, multi-effects scenarios. Europe has been dealing with similar multi-pollutant, multi-effects challenges. While their approach to policy development differs from the US, much can be learned specifically from how the science has supported the development of European policies. For example, European assessments of potential C sequestration in forests include probable enhancement of growth as a result of Nr deposition in N-limited ecosystems, which are quite common. Special attention should be given to the scientific foundations that led to adoption of multiple-pollutant/multiple effects perspectives and the concepts of Critical Loads and Target Loads in management of NO_x, NH_x, VOC, ozone, and PM within the European Union. The Panel has provided a number of references for your consideration in Attachment C.
4. A wide range of “indicators” – including ambient air concentrations (gaseous and aerosol), deposition (wet and dry), and environmental (biogeochemical) – are discussed in the ISA and will later be utilized in the environmental risk and exposure assessments (REA). It would be useful to provide some relevant information in the ISA to illustrate how some of these indicators are related to or distinct from others. Application of these indicators and their relationships should be a focus throughout the document. For example, how do spatial and temporal patterns of gaseous S and N compounds in the ambient air compare to emissions, aerosol concentrations, or deposition?

Responses to EPA’s Charge Questions:

Atmospheric Sciences and Ecological Exposures

- 1. To what extent is the evidence on atmospheric chemistry and physics, air quality, and deposition and exposure sufficiently and correctly described, clearly**

¹ The term total reactive nitrogen includes all biologically, photochemically and radiatively active compounds.

communicated, and relevant to the review of the secondary NAAQS for NO₂ and SO₂?

- 2. How well characterized are the relevant properties of the ambient air concentrations and deposition of NO_x and SO_x, including policy-relevant background concentrations, spatial and temporal patterns, and the relationships between ambient air concentrations and ecological exposures?**
- 3. How sufficient is the information on atmospheric sciences and exposures for the purposes of evaluating and interpreting the ecological effects presented in Chapter 4 of the draft ISA?**

In general, these questions align with Chapters 2 and 3. EPA staff has done a good job identifying the key considerations included in these two chapters and necessary to respond to these charge questions. However, more attention to identifying and quantifying linkages is needed. EPA should also consider combining Chapters 2 and 3 into a single, more integrated chapter. This would effectively present the information in a “source to exposure” format which is more in line with the organization of the Draft ISAs for the Primary NO_x and SO_x standards.

The atmospheric chemistry and physics section was sufficient in coverage and clearly communicated, with the exception of the role of ammonia in gas and particle phase reactions of the NO_x-SO_x-NH_x system. The air quality discussion was hindered by inadequate depictions of the monitoring networks: mixing less relevant networks like ozone with SO₂ and NO₂ networks obscured important information about SO₂/NO₂ monitor location and density. The document concludes that the existing networks are sufficient, but various networks that could contribute data to assess concentrations and deposition each have significant shortcomings. The SO₂/NO₂ monitors are sparse, primarily urban, and absent from large parts of the country. Similar problems plague the particulate matter (PM) speciation network, which is also primarily urban and thus poorly suited to characterize rural and remote areas. Monitoring networks like NADP and CASTNET are critically important for the quantification of deposition and tracking spatial and temporal patterns, but suffer from gaps in coverage as well, and cannot accurately characterize exposure for the broad heterogeneity in sensitive ecosystems. Additional monitoring data exist that ought to be incorporated in this document and used in the risk exposure assessment (e.g., SEARCH). The dearth of ammonia, ammonium and organic Nr data is a significant shortcoming that, while not immediately resolvable, could be addressed by including a summary of data from research studies.

Spatial and temporal trends in emissions and concentrations need to be better represented in figures and/or tables. Emissions of SO_x, NO_x, N₂O and NH_x need to be better quantified, not only at the national level, but also at the regional level. Projections of future emissions should be included in the ISA to put in to perspective the future importance of sulfur oxide and reduced, organic and oxidized nitrogen emissions. Explicit linkages between emissions, ambient concentrations, and deposition should be made. EPA should consider showing maps of SO₂ (and NO_x) emissions adjacent to SO₂ concentrations and SO₄ deposition. The Panel recognizes that this comparison may still be insufficient to provide the quantitative linkages that should ultimately be made.

Air quality modeling appears to be essential to providing quantitative linkages between source emissions and deposition fluxes. The current treatment of model capabilities and accompanying uncertainties in their application should be bolstered. A particular need is to expand discussions of model evaluation and the integration of model results and observations, which are very limited spatially and in terms of species monitored.

Acidification

- 4. How well are the major effects of NO_x and SO_x on ecological acidification identified and characterized? To what extent do the discussions and integration of evidence across scales (e.g. species, communities, ecosystems, and regions) correctly represent and clearly communicate the state of the science?**
- 5. How well has the ISA characterized the relationship between acidifying deposition levels of NO_x and SO_x and environmental effects?**
- 6. How well characterized is the relative importance of the oxidized and the reduced forms of nitrogen on ecosystem acidification?**

The effects of SO_x and NO_x on ecological acidification were adequately identified, characterized, and integrated across scales; however, the Panel has identified modifications or additions for EPA's consideration. A conceptual figure illustrating the highlighted processes could be introduced at the beginning of the document to guide the reader through the entire document. The Agency may consider identifying direct and indirect effects of deposition on acidification. For example, figures showing the direct links – e.g., deposition levels on the x-axis and acidification response(s) on the y-axis – would be helpful. The importance of the SO₄²⁻ and NO₃⁻ mobile anions needs to be highlighted. The Panel would also like the EPA to consider further use of ecosystem functions (production, nutrient cycling, etc.) as response variables to acidifying deposition. The discussion of the natural and anthropogenic processes of soil acidification should be moved to the beginning of the document. Where possible, point out the likely proportions that are due to natural versus anthropogenic causes. Further consideration might also be given to adding a discussion on the effects of capacity (change in soil acidity) and intensity (changes in soil solution, natural water). The former might take decades whereas the latter changes more rapidly in response to the inputs, outputs and cycling of mineral acid anions such as including NO₃⁻, SO₄²⁻ and Cl⁻.

Under indirect effects, the document should note that some important linkages have been made between acidifying deposition and the interaction of other pollutants (e.g., ozone) and other stressors (drought, pests, or pathogens), as well as other processes (fire). Relationships between emission and deposition (or lack of such) should be reiterated here. Reduced and oxidized forms of nitrogen were characterized well; though as previously noted, the influence of all forms of reactive nitrogen on acidification (in addition to NO_x) should be balanced.

Nitrogen Enrichment

- 7. How well are the major effects of NO_x as it contributes to nitrogen enrichment of the ecosystems appropriately identified and characterized? To what extent do the discussions and integration of evidence across scales (e.g. various species, communities, ecosystems, and regions) correctly represent and clearly communicate the state of the science?**
- 8. How well characterized are the relationships between ambient atmospheric nitrogen concentrations, nitrogen deposition and total nitrogen loads, and environmental effects?**
- 9. To what extent has the draft ISA adequately characterized the contribution of oxidized and reduced forms of nitrogen to ecological effects related to nutrient enrichment?**

The impact of nutrient enrichment on terrestrial and aquatic ecosystems is a major focus of this first draft ISA. Although adequate attention is given to nutrient enrichment effects of oxidized forms of reactive nitrogen (Nr), mainly NO_x and N₂O, the treatment of the environmental effects of reduced and organic forms of reactive nitrogen (Nr) is inadequate. Increased emphasis on the wet and dry atmospheric deposition of ammonium ion (NH₄⁺), gaseous ammonia (NH₃) and inorganic Nr is needed in the Second Draft ISA.

The Panel also recommends the further development of the quantitative relationships between atmospheric concentrations of Nr, deposition rates and the resulting effects. To the extent practical, linkages between source(s) and effect(s) should be demonstrated. As noted previously, additional potential environmental impacts from Nr-deposition and the resulting nutrient enrichment should be further discussed, including such effects as increased plant growth leading to carbon sequestration (including enhancement of production in combination with increasing CO₂), enhanced fire danger and increased potential of windthrow. A further indirect effect is that nutrient enrichment will modify the nitrogen cycling: included in this modification is the alteration of N inputs and outputs to/from the system as a result of biotic and abiotic factors such as N fixation by plants and fires.

Additional case studies should be also added to better represent the various U.S. ecosystems and deposition scenarios. Examples for inclusion in the ISA are the mixed conifer forests of the San Bernardino Mountains, coastal sage and desert ecosystems of southern California, which experience high gradients of N deposition.

Other Welfare Effects

- 10. Several additional effects are discussed, including mercury methylation, direct gas-phase effects on foliage, and N₂O as a greenhouse gas. How well does the draft ISA characterize the evidence on these topics?**

Section 4.4 of the report appropriately described available information and discussion of additional NO_x and SO_x deposition-related welfare effects including the link between

atmospheric ambient sulfate and mercury methylation; the direct phytotoxic effects of gaseous nitrogen and sulfur oxides on vegetation (including HNO₃); enhanced production of N₂O from N saturation; and the elevation of nitrate in surface water. The EPA should expand this section to include succinct discussions of the contribution of NO_x and SO_x to the production of particulate matter and tropospheric ozone. Additionally, the Panel recommends that new sections be added on the relationship between nitrogen deposition and carbon sequestration within natural ecosystems and impacts on fires and other potential feedbacks. The discussion should include reasoning as to why certain effects are not more fully considered within the ISA in regards to evaluating as secondary NAAQS for NO_x and SO_x. This might simply be recognition that they are fully considered elsewhere in documents related to other NAAQS. The discussion of methylation should be addressed in the context of the local, regional and global geochemical cycling of both Hg and SO_x and that the two share common sources. Further, to the extent practical, the ISA should indicate the relative NO_x and SO_x contributions to each of the welfare effects listed above (e.g., what fraction of global N₂O emission are attributable to anthropogenic N-enrichment).

Conclusions

11. What are the views of the Panel on the appropriateness and comprehensiveness of the conclusions drawn in Chapter 5?

12. How adequate is the draft ISA for providing information and guidance to future exposure, risk and policy assessments that may be prepared in support of this NAAQS review?

Generally the Panel found that the conclusions, presented in Chapter 5 of the ISA, were logically and appropriately drawn from the information presented in the preceding chapters and annexes. In some cases the concluding material could be more efficiently organized so that there is better congruence between the conclusions and the information presented in the chapters. The Panel noted that the language employed is occasionally more subjective or speculative than necessary. It would be more rigorous to employ a more consistent and liberal use of references to document the concluding statements. The Panel also suggests that a condensed version of the conclusions section be brought forward to the beginning of the ISA document as an executive summary.

While brevity is desirable throughout this ISA document, some of the concluding sections were so pared down that they failed to convey much useful information. For example, the summarization of case studies in Chapter 5 is inadequate to convey much detail about the characteristics of the case study locations, the reasons for their selection, the implications and significance of the environmental effects of S and N deposition on ecosystem resources and functions in these locations, or the intended future analyses in the environmental risk and exposure assessments. Related effects of S and N pollutants – such as on materials damage, visibility and climate forcing – which are not the intended focus of this assessment, need to be briefly summarized here, with pointers to locations where the relevant information can be accessed in other documents.

A key basis for policy decisions is a clear demonstration that introduction of a control strategy (e.g. decreasing Nr and S emissions) will produce the desired decreases in adverse effects. This calls for a demonstration that the existing evidence shows links between emissions, atmospheric concentration, deposition, water and/or soil concentration, and harmful effects on ecosystem resources, services and functions. This chapter should be the place at which this crucial set of causal connections are summarized and judged as to their strength.

The draft ISA provides an appropriate review and summary of the science regarding the effects of S and N deposition on ecosystem chemistry and biology and this will be very helpful for the future risk and policy assessments. However, as noted above, the ISA document would be more helpful if the implications of all effects on ecosystem functions were more clearly articulated. This will provide an essential link to the policy assessment regarding the adversity of these effects.

The degree to which this ISA is going to address economic valuation and ecosystem services needs to be clarified. The ISA should specifically note that linkages between ecosystem functions, changes in ecosystem services and ultimately to articulation of the welfare effects and potential economic valuation will be developed in the subsequent risk and policy assessments. Economic valuation is mentioned in the introduction to the ISA as a potential means to determining what constitutes an adverse effect, but is not mentioned again in the main ISA document, except for a lone table in the conclusions chapter that lists some economic valuation study results and in the Appendix (as a review of the current literature). It is important to clarify that economic valuation is not necessary for an assessment of the adversity of an effect, although if available it would be useful. Further, there was some discussion as to how far this ISA should go in regards to ecosystem function-ecosystem service linkages, versus how much will be covered in later documents (e.g., the REA documents). To the degree the ISA does cover those linkages, it should explore mapping relevant ecosystem functions (e.g., nutrient cycles, system acidification, productivity) that are affected by Nr and SO_x onto specific ecosystem services (ecosystem services as defined by the Millennium Ecosystem Assessment for example). The resulting organizational sequence—from emission to deposition to effects on ecological function to identifying where the affected ecosystem functions fit into ecosystem services—may provide the scientific framework for the development, description and valuation of changes in ecosystem services.

In summary, the information provided in this first draft ISA needs to be further developed to provide the range of information desired to review the secondary NAAQS for NO_x and SO_x. The need for the quantitative linkages between ambient air quality, depositional fluxes, and the resulting effects to the extent practical cannot be overstated. This applies to not only nitrogen and sulfur oxides, but reduced and organic forms of nitrogen as well.

The CASAC NO_x & SO_x Secondary NAAQS Review Panel was pleased to review the first draft of the *ISA for Oxides of Nitrogen and Sulfur – Environmental Criteria*. We look forward to review of the second draft and wish you well in this important endeavor.

Sincerely,

/Signed/

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

/Signed/

Dr. Rogene Henderson, Chair
Clean Air Scientific Advisory Committee

Enclosures

Appendix A: Roster of CASAC NO_x & SO_x Secondary NAAQS Review Panel

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

CASAC MEMBERS

Dr. Armistead (Ted) Russell (*Chair*), Professor, Department of Civil and Environmental Engineering, Georgia Institute of Technology, Atlanta, GA

Dr. Ellis B. Cowling, University Distinguished Professor At-Large Emeritus, Colleges of Natural Resources and Agriculture and Life Sciences, North Carolina State University, Raleigh, NC

Dr. Douglas Crawford-Brown, Professor Emeritus and Director Emeritus, Department of Environmental Sciences and Engineering and UNC Institute for the Environment, University of North Carolina at Chapel Hill, Chapel Hill, NC

Dr. Donna Kenski, Data Analysis Director, Lake Michigan Air Directors Consortium, Rosemont, IL

PANEL MEMBERS

Dr. Praveen Amar, Director, Science and Policy, NESCAUM, Boston, MA

Dr. Andrzej Bytnerowicz, Senior Scientist, Pacific Southwest Research Station, USDA Forest Service, Riverside, CA

Ms. Lauraine Chestnut, Managing Economist, Stratus Consulting Inc., Boulder, CO

Dr. Charles T. Driscoll, Jr., Professor, Environmental Systems Engineering, College of Engineering and Computer Science, Syracuse University, Syracuse, NY

Dr. Paul J. Hanson, Distinguished R&D Staff Member, Environmental Sciences Division, Oak Ridge National Laboratory, Oak Ridge, TN

Dr. Rudolf Husar, Professor and Director, Mechanical Engineering, Engineering and Applied Science, Center for Air Pollution Impact & Trend Analysis (CAPITA), Washington University, St. Louis, MO

Dr. Dale Johnson, Professor, Department of Environmental and Resource Sciences, College of Agriculture, University of Nevada, Reno, NV

Dr. Naresh Kumar, Senior Program Manager, Environment Division, Electric Power Research Institute, Palo Alto, CA

Dr. Myron Mitchell, Distinguished Professor and Director of Council on Hydrologic Systems Science, College of Environmental and Forestry, State University of New York, Syracuse, NY

Mr. Richard L. Poirot, Environmental Analyst, Air Pollution Control Division, Department of Environmental Conservation, Vermont Agency of Natural Resources, Waterbury, VT

Mr. David J. Shaw, Director, Division of Air Resources, New York State Department of Environmental Conservation, Albany, NY

Dr. Kathleen Weathers, Senior Scientist, Institute of Ecosystem Studies, Millbrook, NY

SCIENCE ADVISORY BOARD STAFF

Ms. Kyndall Barry, Designated Federal Officer, 1200 Pennsylvania Avenue, NW
1400F, Washington, DC, Phone: 202-343-9868, Fax: 202-233-0643, (barry.kyndall@epa.gov)

Appendix B: Roster of the chartered CASAC

**U.S. Environmental Protection Agency
Science Advisory Board (SAB) Staff Office
Clean Air Scientific Advisory Committee (CASAC)**

CASAC MEMBERS

Dr. Rogene Henderson (*Chair*), Scientist Emeritus, Lovelace Respiratory Research Institute, Albuquerque, NM

Dr. Ellis B. Cowling, University Distinguished Professor At-Large Emeritus, Colleges of Natural Resources and Agriculture and Life Sciences, North Carolina State University, Raleigh, NC

Dr. James D. Crapo, Professor, Department of Medicine, National Jewish Medical and Research Center, Denver, CO

Dr. Douglas Crawford-Brown, Professor Emeritus and Director Emeritus, Department of Environmental Sciences and Engineering and UNC Institute for the Environment, University of North Carolina at Chapel Hill, Chapel Hill, NC

Dr. Donna Kenski, Data Analysis Director, Lake Michigan Air Directors Consortium, Rosemont, IL

Dr. Armistead (Ted) Russell, Professor, Department of Civil and Environmental Engineering, Georgia Institute of Technology, Atlanta, GA

Dr. Jonathan Samet, Professor and Chairman, Department of Epidemiology, Bloomberg School of Public Health, Johns Hopkins University, Baltimore, MD

SCIENCE ADVISORY BOARD STAFF

Mr. Fred Butterfield, CASAC Designated Federal Officer, 1200 Pennsylvania Avenue, N.W., Washington, DC, 20460, Phone: 202-343-9994, Fax: 202-233-0643, (butterfield.fred@epa.gov)

Appendix C: Compilation of Individual Panel Member Comments on EPA’s *Integrated Science Assessment (ISA) for Oxides of Nitrogen and Sulfur – Environmental Criteria* (December 2007)

Comments received:

Dr. Praveen Amar	13
Dr. Andrzej Bytnerowicz	18
Ms. Lauraine Chestnut	24
Dr. Ellis B. Cowling	26
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Dr. Praveen Amar

This write up addresses the first three charge questions from NCEA. The Charge Questions are reproduced below:

- 1. To what extent is the evidence on atmospheric chemistry and physics, air quality, and deposition and exposure sufficiently and correctly described, clearly communicated, and relevant to the review of the secondary NAAQS for NO₂ and SO₂?*
- 2. How well characterized are the relevant properties of the ambient air concentrations and deposition of NO_x and SO_x, including policy-relevant background concentrations, spatial and temporal patterns, and the relationships between ambient air concentrations and ecological exposure?*
- 3. How sufficient is the information on atmospheric sciences and exposures for the purposes of evaluating and interpreting the ecological effects presented in Chapter Four of the draft ISA?*

Response:

Chapter 2 (The Atmospheric Chemistry and Physics of Nitrogen and Sulfur Oxides, as well as Annex AX1 with the same Title), and Chapter 3 (Ecological Exposure to Oxides of Nitrogen and Sulfur, and to Ammonia and ammonium, as well as Annex AX2 with the same Title) provide a reasonably detailed description of the atmospheric science, air quality, deposition, and exposure. However, the Chapters can and should be improved for more clear communication. It is understandable that various sections have different authorships. These Chapters and other chapters in the ISA would therefore benefit from the services of an expert technical/scientific editor resulting in a more readable ISA that more clearly communicates the important findings of these Chapters (as well as other chapters).

Specific Comments to improve the ISA are provided below:

Page 2-2: Line 8: It is important to state the more important point first, followed by a minor point. Ammonia is included in this ISA, first because of its role in NO_x and SO₂ chemistry, and its role in nitrification, and, *second* (and a distant second!), because its oxidation can be a minor source of NO_x. There are other parts of this ISA that do suffer from the same issue. That is, first order issues should be mentioned first.

A General comment about time scales of various chemicals/reactions: The ISA does a very good job of providing time scales of various reactions and species. However, it would be very useful if many of these important reactions' time scales are presented in a single Table, where readers can compare them in one place and draw meaningful conclusions.

Page 2-4: Line 17: P (O₃): Ozone *Production* Efficiency: It is an important concept. However, I do not think it is clearly defined in the document (as number of molecules of ozone produced per

molecule of NO_x over a certain time period and over a spatial extent, etc.). I suggest it should be explicitly defined first time it appears.

Page 2-7 (Lines 22-23): Awkward and an out-of-context reference to the fact that NO₃ deposition is a “complex function of wind speed.” What does this mean? Needs an explanation.

General Comment about Section 2.2.2: Halogen Chemistry in the Marine Layer: This section is written rather poorly and seems to be out of context with the main theme of SO₂ and NO_x chemistry. I found it hard to follow. If it has important implications for the deposition of nitrogen and sulfur compounds on coastal and non-coastal/regional scales, they should be stated.

Page 2-9, General Sulfur Chemistry: Besides SO₂, SO₃ though small in amount compared to SO₂ (about 1 to 3 percent of total emitted SO_x in stacks) has been known to cause visibility degradation (“Blue plume” downwind of large power plants burning high sulfur coal, and equipped with pollution controls of SCR and FGDs). It needs to be addressed here or in the PM document.

Page 2-10: Line 1: It is stated that aqueous-phase oxidation of SO₂ is responsible for about 80% of the total oxidation (implying that the remaining 20 percent is by homogeneous gas phase oxidation or by metal-catalyzed reactions). However, the ISA should note that the situation is lot more complex than that. For example, which of the various oxidation pathways dominates depends on the details of the local accumulation of the co-pollutants such as ozone, hydrogen peroxide, hydrocarbons, ammonia, and catalytic metals (Fe, Mn, and others). This will have an impact on final deposition levels of sulfur.

Page 2-14: A clear explanation of S (IV) and S (VI) would be helpful.

Page 2-21: Section on Satellites: A comment similar to the comment on Halogen Chemistry above. This section appears to have been written out-of-context and needs to be integrated with the larger context of estimating ecological exposures to S and N deposition and how satellite data may be helpful in providing total loadings of S and N on larger scales that only satellites can provide. Same comment on Table 2.6-1. May be, the Table needs to be removed.

Page 3.2: SO_x Emissions: Lines 14-18: The statement that sources other than electric utilities “make only a very minor contribution” to overall SO₂ emissions is quite inaccurate and needs to be corrected. In 2003, the non-EGU sources in the US contributed 31% of the total SO₂ emissions (at 5 mm tpy compared to about 11 mm tpy for EGUs). For example, industrial, commercial, and institutional (ICI) boilers burning coal and oil produce about one mm tpy of SO₂ in the US. Same text appears in the Annex and needs to be corrected. Also, the Summary (page 3-59) needs to be corrected.

Page 3.9 NH₃ Emissions, Lines 23-28: Very awkward description of ammonia emissions. Three-Way catalysts and ammonia emission from them are NOT the primary source of ammonia, and therefore, a description of ammonia emissions must start with known large sources of ammonia (livestock and agricultural operations). Please rewrite for more effective communication and drop the words “for these reasons.. .”

A potentially important point about ammonia emissions is the geographic shift in ammonia emissions that seems to have taken place in the US between 1980s and late 1990s and may still be accelerating. The attached slide, courtesy of Dr. Bruce Hicks, NOAA, shows that the “center of ammonia emissions” may be shifting from Midwest to the Southeast and the Carolinas. That is, ammonia emissions (in N kg/ha) have increased by about 2 to 3 Kg/ha in the Southeast and gone down by 1 to 2 kg/ha in the Midwest. This shift in density of ammonia emissions has obvious implications on the regional atmospheric chemistry of SO_x, NO_x, and ammonia and the resulting regional wet and dry deposition patterns for both sulfate and nitrates. ISA should address the impact of this potential trend in geographic change in ammonia emissions.

Page 3-10: Section 3.4: Evaluating Emission Inventories: The ISA should note that the first step to evaluate emission inventories is not by looking at the ambient concentrations. The first step in evaluating reliability and accuracy of emission inventories needs to take into account the methods and procedures that are used to produce emission inventories themselves (use of emission factors, use of CEMs, QA/QC methods, uncertainty in determining spatial and temporal profiles, speciation factors for VOCs and primary PM emissions, etc.).

Page 3-11, Section 3.5.2 NO₂: Say “NO₂ interaction with vegetation is MORE DIFFICULT TO UNDERSTAND than...”

Page 3-16 Line 10: Not clear about the distinction between “aerosol-phase” and “solid-phase”. Are they not the same in the context of atmospheric chemistry of aerosols?

Page 3-16, Line 7: “Title IV Reductions in NO_x and SO₂” and not N and S.

Page 3-17, Figure 3.6-3: The Title needs to be modified to indicate that the Figure also shows trends, if any (2002 to 2006).

Page 3-19, Lines 8-10: Please rewrite for clarity.

Page 3-21: Satellites: same comment as above. Please note the larger context of S and N deposition.

Page 3-34: Section 3-9: Harvard Forest: I have a comment similar to the comment on Halogen Chemistry above. It reads as an independent piece without being integrated into the ISA. It is too long and not clear. Please shorten it and rewrite.

Page 3.12 PRB Concentrations of NO_x and SO_x: This is well-written section and does a very good job of responding to the Charge Question 2 on the subject of PRBs of NO₂, SO₂, and nitrate and sulfate deposition over various regions in the US and at different time scales.

Page 3-43 Regional-Scale CTMs, Also Annex AX-2 (AX2.10.1.1, Page AX2-62): The text notes that the capabilities of a number of CTMs designed to study local and regional-scale air pollution problems were summarized by Russell and Dennis (2000) and by others, including Arnold (2003), etc. What is missing is the reference to recent peer reviews of the CMAQ system itself.

The ISA needs to recognize that CMAQ modeling system has been extensively peer-reviewed. The ISA document should refer in Chapter 3 and in Annex AX-2 to the three detailed written reports of the external peer review panels. It would also be useful to include a short summary of the major findings of the peer review reports that were published by the CMAS in years 2003, 2005, and 2007. They are available on CMAS website (www.cmascenter.org). The three references (they should be included both in Chapter 3 and Annex AX-2) are:

1. Amar, P., R. Bornstein, H. Feldman, H. Jeffries, D Steyn, R. Yamartino and Y. Zhang, 2004: *Final report: December 2003 Peer Review of the CMAQ Model*. Report submitted to CMAS Center, University of North Carolina at Chapel Hill, July, 24 pp.
2. Amar, P., D. Chock, A. Hansen, M. Moran, A. Russell, D. Steyn and W. Stockwell, 2005: *Final Report: Second Peer Review of the CMAQ Model*. Report submitted to CMAS Center, University of North Carolina at Chapel Hill, July, 33 pp.
3. Aiyyer, A., D. Cohan, A. Russell, W. Stockwell, S. Tanrikulu, W. Vizuete and J. Wilczak, 2007: *Final report: Third Peer Review of the CMAQ Model*. Report submitted to CMAS Center, University of North Carolina at Chapel Hill, February, 25 pp.

A General Comment on Dry Deposition: The ISA needs to be more clear and explicit that we only ESTIMATE dry deposition and therefore comments about total deposition (wet and dry) and about the relative contribution of each pathway have a level of uncertainty that is hard to determine.

In summary, Chapters 2 and 3 and annexes address the first three charge questions on atmospheric science and ecological exposure in a satisfactory manner. However, the whole document needs a thorough technical and scientific editing job as well as a better integration of some of the sections (Halogen Chemistry, Harvard Forest, satellites use) into the overall ISA.

Chapter 5: Findings and Conclusions

General Comment: This Chapter does a good job of summarizing the ISA with major “take home” messages. As noted above for Chapter 2 and 3, this Chapter does need a thorough scientific editing job for it to communicate complex concepts in a clear manner.

Specific Comments follow:

Page 5-2 Section 5.2.2: The conclusion drawn here that current routine monitoring is “adequate” or “fully adequate” after listing major problems with measurement of gaseous and particulate species does not seem to be justified. I think it is a matter of tone and the revised language should indicate WHY the current network is adequate.

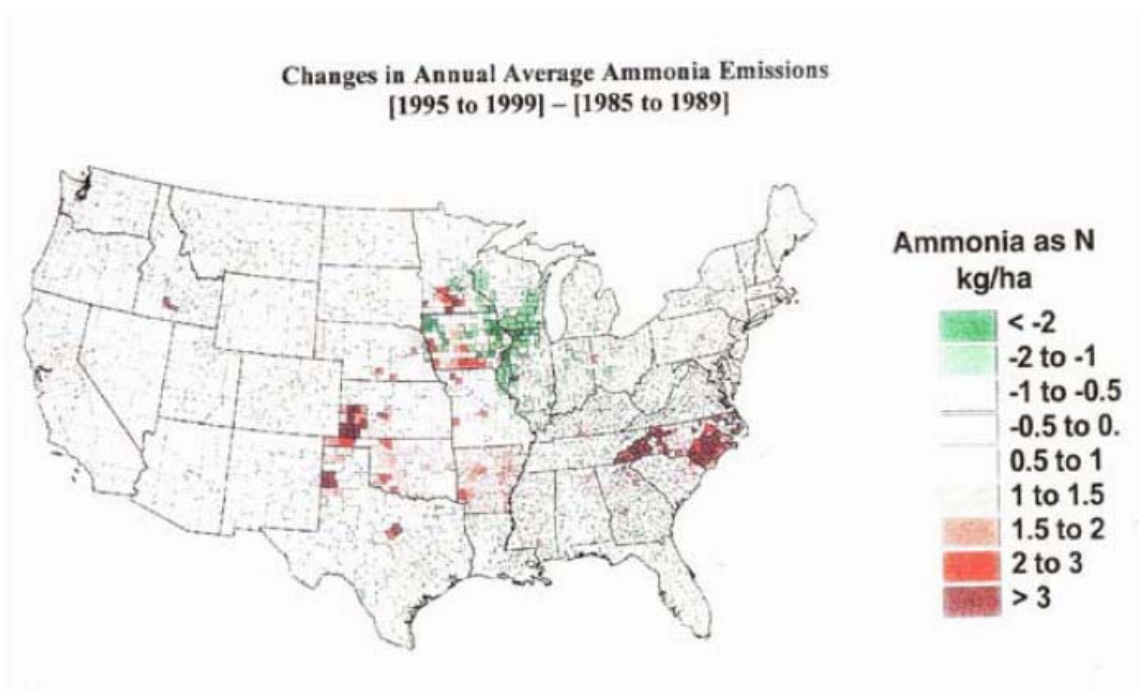
Page 5-3 Line 30: Please see the comments for Chapter 2 about SO₂ emissions from sources other than power plants. They are not “very little,” since they are about 1/3 of the total SO₂ emissions.

Page 5-11: Lines 20-31: Not clear why there is no relationship between recent trend in N deposition and trends in nitrate concentrations in surface waters. Have the recent reductions in NO_x emissions in the eastern US because of the “NO_x SIP Call” taken into account?

Page 5-36: Line 20: Please see my comment above on “adequate monitoring.” Here, it says that the monitoring networks are “inadequate.” Need to be consistent.

Geographic Shift in Ammonia Emissions

(courtesy of Bruce Hicks, NOAA)



Dr. Andrzej Bytnerowicz

General Comments:

Chapter 4 is well written and comprehensive and presents state of the scientific knowledge on ecological effects of acidification, nitrogen nutrient additions and other welfare effects of N and S. The chapter is very informative and is based on the relevant and current peer-reviewed literature from North America and Europe.

Having said this, I would like to suggest that N effects are presented in a context of multi-pollutant, multi-effects scenarios. Example of such an approach is the 1999 Gothenburg protocol of the UN Convention of Long-range Transport of Air Pollutants recommending evaluation of the combined effects of NO_x, SO_x, NH₃, and VOCs on acidification, eutrophication and ground levels ozone impacts (Working Group on Effects, 2004).

Specific comments:

Page 4-4, Figure 4.1.-1. should be changed to Table 4.1.-1.

Why eutrophication is not shown as a possible disturbance if forests?

In row 7 (atmospheric pollutant), the end products of deposition are shown, but not the pollutants causing biological effects (e.g., NO₂, SO₂, HNO₃, NH₃, particulate NO₃, particulate NH₄).

Page 4-5, point 7 – ammonia (NH₃) should be listed as well.

Page 4-9, Figure 4.2-1 mixed up with Fig. 4.2-2.

Page 4-13, lines 12-27. Elevated levels of tropospheric ozone and land disturbances such as catastrophic fires or withdrthrows should be mentioned as factors affecting water resources of ecosystems and their leaching potential.

Page 4-22, lines 25-27. This could be a good recommendation for future research needs.

Page 4-29, lines 27-28. Reduced cold tolerance is also caused by eutrophication.

Page 4-35, line 14. These effects on epiphytes, if I recall, were also caused by NO₂ and SO₂.

Page 4-35, section 4.2.2.2.3. The described effects are to a large extent not caused by acidity but direct toxic effects of SO₂ on plants.

Pages 4-38 through 4-44. Change SO₄⁺ to SO₄²⁻.

Page 4-66, lines 26-28. Reference is needed.

Page 4-73, lines 28-32. It is a very important statement – clearly no single “definitive” critical load for US ecosystems is possible. The eco-regional approach to the CL issue seems to make most sense.

Page 4-98, lines 15-17. Is it contribution of gaseous HNO₃ or of total dissolved NO₃⁻?

Page 4-121, lines 18-20. In regard to changes in lichen species composition, there are differences in responses to reduced vs. oxidized N. It has been proposed that elevated levels of HNO₃ have negative effects on sensitive lichens such as *Ramalina menziesii* in the San Bernardino Mountains of southern California (Riddell et al., 2008). Such effects are attributed more to the direct HNO₃ toxicity than NO₃⁻ deposition. There is also a high probability that the reduced N resulting from high NH₃ and particulate NH₄⁺ could affect lichens downwind of the Los Angeles pollution sources area (Prof. Eva Barreno, personal communication). However, it has to be remembered that the disappearance of lichens in southern California could also be caused by high levels of ambient ozone in the area (Nash and Sigal, 1998; Sigal and Nash, 1983).

Page 4-122, lines 20-26. It should be added that increased mortality of the high elevation trees in areas of elevated N deposition could be partially caused by their increased sensitivity to frost.

Page 4-129, section 4.3.3.1.4. This section should be incorporated into the forests section (4.3.3.1.1) (see my remarks below).

Page 4-131, section 4.3.3.1.6. This section should be divided and moved into “forests” and “arid & semi-arid” ecosystems (see my remarks below).

Page 4-147, lines 28-30. Transitional ecosystems (as mentioned in the previous sentences) should also be added.

Page 4-152, lines 15-18. I do not see a logical link here. If estuaries and coastal water are inherently sensitive to increased N loading, this is not because of the high releases of N inputs back to the atmosphere (high microbial activity). Increased release of N into the atmosphere can be considered as an avoidance mechanism in the presence of excessive N loads.

Page 4-154, lines 21-24. Ranges of deposition that may be affecting sensitive species seem to be mixed up – levels in alpine ecosystems should be lower than in other terrestrial ecosystems cited in this ISA document (see Baron et al. 2000; Bowman et al., 2006).

Page 4-162, line 8. S deficiency level at <80 µg/g seems to be very low. In Scots pine foliage in the pristine area of Europe the lowest recorded level of S was ~400 µg/g (Molski et al., 1981).

Page 4-176. Section on the direct effects of HNO₃ on plants should be considered and possibly added.

Page 4-180, Table 4.2-2. Add “of” after “Example”.

Page 4-186, Table 4.2-9. Change “eith” to “with” in the second line.

Page 4-189. Table 4.3-2. in the column “Factors that govern vulnerability”, add a sentence “Interactions with other contributing stressors such as elevated levels of ozone or drought”.

Replies to the assigned questions:

7A. How well are the major effects of NO_x as it contributes to nitrogen enrichment of the ecosystems appropriately identified and characterized?

Generally the effects of NO_x contribution to the N enrichment of various ecosystems are well described. These effects are divided into the terrestrial, transitional, aquatic (fresh water and estuarine and coastal water) ecosystems. Two case studies (alpine and sub-alpine communities in Colorado and in Chesapeake Bay) are described. Some other case studies could be possibly added – such as the mixed conifer forest of the San Bernardino Mountains (Fenn and Poth, 1999; 2001; Fenn et al., 2003) and grassland ecosystems in the San Francisco Bay area (Weiss, 1999).

7B. To what extent do the discussions and integration of evidence across scales (e.g.. various species, communities, ecosystems, and regions) correctly represent and clearly communicate the state of science?

Generally the effects (biogeochemical processes and biological effects) have been described (as in point 1) for the terrestrial, transitional and aquatic ecosystems from a perspective of the effects indicators. Some confusion is caused by placing herbaceous and plants and shrubs (4.3.3.1.4) and mycorrhizal and microbial activity (4.3.3.1.6) into separate sections. Information contained in those sections should be incorporated into the forests (4.3.3.1.1) and arid and semi-arid ecosystems (4.3.3.1.3). Lichens could stay alone since their evaluation surveys have been done mostly for larger geographic areas encompassing various ecosystems (often including agricultural or the urban-wildland interface). What is missing are the desert ecosystems – effects of N deposition in these N-limited systems, especially in the vicinity of strong N sources areas, should be mentioned. A good example is the Mojave Desert in California which is strongly affected by N deposition from the Los Angeles area. There are only a few references referring to those problems, however, research is ongoing at the University of California in Riverside (Allen et al., 2008).

8. How well characterized are the relationships between ambient atmospheric nitrogen concentrations, nitrogen deposition and total nitrogen loads, and environmental effects?

These relationships are not well described. This is mainly caused by a lack of sufficient understanding of these relationships. Understanding responses of ecosystems to N addition are often based on fertilization experiments, using mainly NH₄NO₃ or (NH₄)₂SO₄ additions and assuming that these additions would simulate wide spectra of atmospheric N deposition scenarios. More experiments on natural N deposition gradients, with the adequately identified N components (gases, aerosols and soluble ions), should be conducted. Deposition of N to forest ecosystems is calculated mostly from throughfall data with an assumption that wet and dry deposition components are included. This approach has been used at large scales in Europe and helped in calculating critical loads for N and S deposition and acidity (ICP Forests and ICP Maps

and Modeling products). However, the N dry deposition component, especially when there is not sufficient precipitation to remove N deposited on foliage and other surfaces to throughfall solution, may be grossly miscalculated. The inferential method (using information on ambient concentrations of major N deposition drivers, their deposition velocity as well as canopy characteristics, such as leaf area index, LAI) may be helpful in such cases. This method could also be applied in such ecosystems (grasslands, deserts, alpine and sub-alpine ecosystems) where throughfall cannot be used at all or is very difficult to be applied. The inferential method has numerous limitations and uncertainties; however, it can be used for large geographic areas. This method is the basis for the CASTNET estimates of N dry deposition at the national scale. It has to be remembered, however, that there is not enough data for ambient NH_3 , which is one of the main drivers of N dry deposition. In addition, understanding of N deposition in complex (mountain) terrains is still poor.

9. To what extent has the draft ISA adequately characterized the contribution of oxidized and reduced forms of nitrogen to ecological effects related to nutrient enrichment?

Contribution of reduced vs. oxidized forms of atmospheric nitrogen to nutrient enrichment effects are not well understood and subsequently not well described in the ISA. The main reason for that is an inadequate understanding of the chemical environment of ecosystems for which the effects of N deposition are being described. As I mentioned above, most of knowledge on the ecosystem responses comes from the fertilizer studies. Such experiments are well suited for understanding biological effects of N in the wet deposition dominated ecosystems, such forests in the eastern United States. However, in the arid and semi-arid ecosystems, where large portion of N deposition may result from gaseous HNO_3 , NH_3 or particulate NO_3^- or NH_4^+ , fertilization experiments may quite poorly mimic the real-world interactions between the atmosphere and ecosystems (vegetation, soils and surface water components).

Recommendations:

1. While SO_2 and NO_x concentrations should be monitored for health reason (primary standards), they have very limited application for evaluation of environmental (ecological) effects. Therefore there is a need for monitoring concentrations of major drivers of N & S deposition. For S, this is mostly SO_4 in wet precipitation measured nationally (e.g. NADP network). However, for N, not only wet NO_3 and NH_4 deposition (also measured nationally), but also dry deposition of gaseous HNO_3 , NH_3 and particulate NO_3 and NH_4 as well as the organic compounds should be included. Activities of the national monitoring networks, especially CASTNet should be continued and expanded, especially in the underrepresented Western US.
2. For preliminary determination of HNO_3 and NH_3 “hot spots”, use of passive samplers should be considered. Passive samplers have already been successfully used in monitoring networks in California: Sequoia National Park (Bytnerowicz et al., 2003), Lake Tahoe (Gertler et al., 2006), or Joshua Tree National Park (Allen et al., 2008).
3. National efforts in ecological monitoring of N & S deposition effects are needed. Such efforts could be similar to those in Europe under the auspices of the UN Economical Commission for Europe, namely the ICP Forests Level and ICP Modelling and Mapping efforts. Monitoring results from the ICP Forests Level II plots have allowed for

determination of CL and their exceedances. The newly developed network for CL estimates on 17 US Forest Service Experimental Forests and additional 4 sites across the US will help to determine if the CL approach could be applied and be practical in the US. Research collaboration within the US and with the Canadian and European partners is planned. If successful, the monitoring efforts in the US FS could be expanded into a denser national network. It is envisioned that US FS Forest Inventory and Analysis (FIA) Phase 3 (Forest Health Monitoring) sites could be utilized. The Intensive Site Monitoring (ISM) site that would integrate the FIA Phase 3 and the ICP Forests Level II-type plots will be established in the San Bernardino Mountains of southern California in summer 2008.

4. Effective collaboration between EPA, various national monitoring networks, and land management agencies such as US Forest Service or National Park Service, in developing a national program for monitoring and evaluation of N & S effects on ecosystems, including CL calculations, should be encouraged.

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- Weiss SB. 1999. Cars, cows, and checkerspot butterflies: Nitrogen deposition and management of nutrient-poor grasslands for a threatened species. *Conservation Biology* 13, 1476-1486.

Some literature citations supporting evidence of increased windthrow in forests experiencing elevated N deposition:

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Ms. Lauraine Chestnut

Charge question 11 (appropriateness and comprehensiveness of conclusions?)

The review of relevant economic valuation studies is included only as an appendix and is not incorporated into the conclusions. Table 5.5-2 lists some results of water quality valuation studies in the Chesapeake Bay, but I don't see discussion of this table in the text. As a result of both of these factors, the treatment of the economic valuation literature seems disconnected from the review of the science. It is not clear how the review of the economics literature contributes and fits into this ISA. There was mention in the introduction that economic valuation may be helpful in assessing what effects are adverse, but I don't see any discussion of specifically how the current literature may be useful in this regard in the appendix or in the findings and conclusions. If the ISA is not going to make the link between harmful effects on ecosystem functions to the question of how these translate into welfare effects and associated economic values, then this should be explained in the introduction. This link needs to be made at some point in this process, but it may be better addressed in the risk and policy assessments. If this is EPA's intention, it needs to be stated in the introduction. The ongoing work of the SAB Committee on Valuing the Protection of Ecological Systems and Services (C-VPES) and EPA's Ecological Benefits Assessment Strategic Plan should also be cited here as important background materials for economic valuation methods.

It is also important to stress that a determination of there being an adverse welfare effect does not necessarily require that it be quantified in monetary value terms. Economic valuation studies may be helpful in assessing when an affect is adverse, especially for direct use values. However, for total value (including nonuse, bequest, and even some indirect use values) the public can only assess these values when they are fully informed about what the changes to the ecosystem resources and functions are, including an understanding of the implications of these changes on the quality of the many ecosystem services that these resources provide. This information and understanding has to have its foundations in the science. The conclusions in the ISA would be more useful if they articulated the implications of the changes observed in the various ecosystems affected by N and S deposition in terms of changes or losses in ecosystem functions.

Charge question 12 (adequate information and guidance for the exposure, risk and policy assessments?)

In reviewing the ISA I kept looking for two types of information that seem to be key to this process of considering secondary standards:

1. What information is there that tells us when a change in an ecosystem might be considered adverse?
2. What levels of deposition/exposure can a system tolerate without incurring adverse changes?

Perhaps these questions are to be addressed more explicitly in the risk assessment phase. I didn't see them explicitly addressed in the ISA. A few observations related to the question of what exposures can be tolerated include:

1. Exposures that can be tolerated vary for different resources and different locations according to several known characteristics.
2. It will be very hard to say what levels of N deposition can be tolerated when there are significant other sources in aquatic systems (especially for estuaries).
3. The answer may be different for recovery of currently injured resources than for prevention of future degradation.
4. The question is complicated by episodic events (e.g. spring runoff) that cause temporary but damaging increases in exposures.

The ISA notes that the current secondary standards for SO₂ and NO₂ were not set to address the effects of deposition, but has the case been made that these standards are not sufficiently protective? Perhaps this is obvious, but if current standards are being met and adverse affects are still occurring, then the standards are not adequate. Related to this issue is the question of whether adverse effects of deposition will continue once the primary NAAQS for PM and ozone are met. These standards are going to require further reductions in SO₂ and NO_x emissions from current levels. At what point in this process does it need to be assessed whether the reductions in deposition that will result will be sufficient to protect ecosystem resources, or is this not relevant?

Dr. Ellis B. Cowling

Individual Comments on the First External Review Draft of the Integrated Science Assessment for the Secondary National Air Quality Standards for Nitrogen Dioxide and Sulfur Dioxide

These Individual Comments are developed in the form of responses to most of the 12 Charge Questions received from Kyndall Barry together with the final agenda for our CASAC Peer Review on April 2, 2008.

Chairman Ted Russell asked Andrzej Bytnerowicz and me to give special attention to Charge Questions 7-9 in the attached list. Thus I am very much looking forward to comparing notes with Andrej during our Peer review on April 2, 2008.

My responses to most of the Charge Question are presented below in ordinary type after presentation of the Charge Questions in **Bold Type**.

1. To what extent is the evidence on atmospheric chemistry and physics, air quality, and deposition and exposure sufficiently and correctly described, clearly communicated, and relevant to the review of the secondary NAAQS for NO₂ and SO₂?

Chapter 2 of this ISA gives a very thorough account of the major sources, and the chemical and physical transport, transformation, and atmospheric deposition processes for oxides of nitrogen and sulfur. The schematic diagram of the cycle of reactive nitrogen in Figure 2.2.1 gives a reasonably firm foundation for understanding those parts of the nitrogen cycle that involve oxidized forms of nitrogen.

But this schematic diagram includes no mention or pictorial illustrations of the huge (mostly agricultural) sources of reduced and organic forms of reactive nitrogen that are critical to understanding both the nitrogen enrichment and acidification effects of atmospherically deposited nitrogen and sulfur on terrestrial and aquatic ecosystems.

If EPA is serious about dealing with both nutrient enrichment and acidification of ecosystems induced by atmospheric deposition of total reactive nitrogen, the Agency needs to include in Chapter 2 of the Second Review Draft of this ISA for "NO_x" and "SO_x," a similar schematic diagram for the major sources, transformation, transport and atmospheric deposition processes for reduced and organic forms of reactive nitrogen.

2. How well characterized are the relevant properties of the ambient air concentrations and deposition of NO_x and SO_x, including policy-relevant background concentrations, spatial and temporal patterns, and the relationships between ambient air concentrations and ecological exposures?

3. How sufficient is the information on atmospheric sciences and exposures for the purposes of evaluating and interpreting the ecological effects presented in Chapter 4 of the draft ISA?

4. How well are the major effects of NO_x and SO_x on ecological acidification identified and characterized? To what extent do the discussions and integration of evidence across scales (e.g. communities, ecosystems, and regions) correctly represent and clearly communicate the state of the science?

The presentation of current scientific knowledge about the effects of NO_x, SO_x, and total reactive nitrogen in Chapter 4 of this First Draft ISA is very thorough and comprehensive. The discussion and integration of evidence across scales (communities, ecosystems, and regions) is also reasonably complete.

5. How well has the ISA characterized the relationship between acidifying deposition levels of NO_x and SO_x and environmental effects?

6. How well characterized is the relative importance of the oxidized and the reduced forms of nitrogen on ecosystem acidification?

Although Chapter 4 of this First Draft ISA gives a very thorough and wide ranging account of the huge increase in scientific understanding of nitrogen enrichment and acidification effects of atmospherically deposited nitrogen and sulfur on the structure, specific components, functions, and ecosystem services provided by many different ecosystems in many different parts of this country, the attention given to comparison of the extent to which these many adverse effects are caused by atmospheric deposition of oxidized and organic forms of nitrogen vs oxidized forms of nitrogen is vanishingly small.

The truth is that although there are short-term differences in the rapidity and specific species effects of nitrogen enrichment effects induced by reduced vs oxidized forms of nitrogen, the long-term effects are essentially indistinguishable.

Thus, the most important policy relevant decision that needs to be made with regard to the Second Draft ISA is to set the stage more adequately for consideration of a “Total Reactive Nitrogen” approach in air quality management in this country and to refrain from trying to deal only with oxidized forms of nitrogen rather than the sum of oxidized, plus reduced, plus organic forms of reactive nitrogen.

7. How well are the major effects of NO_x as it contributes to nitrogen enrichment of the ecosystems appropriately identified and characterized? To what extent do the discussions and integration of evidence across scales (e.g. various species, communities, ecosystems, and regions) correctly represent and clearly communicate the state of the science?

The adverse effects of atmospherically deposited nitrogen and sulfur on terrestrial and aquatic ecosystems fall into two major categories that are reasonably well described in this First Draft Integrated Science Assessment for Oxides of Nitrogen and Sulfur – “Nutrient Enrichment” and “Acidification.” Total reactive nitrogen deposition is the principal cause of

nutrient enrichment whereas both nitrogen and sulfur deposition are the principal causes of acidification in both terrestrial and aquatic ecosystems.

Notably, however, in the context of this CASAC Peer Review of the secondary standards for nitrogen and sulfur oxides, this First Draft ISA fails to demonstrate that it is not just oxidized forms of nitrogen and sulfur that induce major adverse ecosystem effects, but also chemically reduced and organic forms of nitrogen, and to a lesser extent, also reduced and organic forms of sulfur. In some very important parts of the US, atmospheric deposition of reduced and organic forms of nitrogen are larger in total load of reactive nitrogen than the total load of oxidized forms of nitrogen.

The principal sources of oxidized airborne nitrogen and sulfur are combustion of fossil fuels used in production of electricity, industrial processes of many sorts, transportation vehicles, and both commercial and domestic home and water heating systems. These matters are adequately covered in this First Draft ISA. But this is NOT true for the very important reduced and organic forms of nitrogen.

The principal sources of air emissions of reduced and organic forms of nitrogen are agricultural operations that include fertilization of crops and forests, but even more importantly include animal rearing operations (principally chicken, turkey, and egg production units, swine farms, and both beef and dairy cattle farms) but also including other domestic animals such as horses, goats, sheep, and even companion animals. Municipal waste handling and processing facilities and septic tank systems are also important sources of air emissions of ammonia and ammonium ion.

Please see the comment written in response to the first Charge Question listed above with regard to the need for a schematic diagram similar to Figure 2.1.1, but developed for reduced and oxidized forms of nitrogen rather than just for oxidized forms of nitrogen.

8. How well characterized are the relationships between ambient atmospheric nitrogen concentrations, nitrogen deposition and total nitrogen loads, and environmental effects?

My impression is that the contributions of inorganic forms of nitrogen (principally nitrate ions and ammonium ion concentrations to total nitrogen loads in wet deposition are reasonably well characterized. On the other hand, the contribution of organic forms of nitrogen to total reactive nitrogen loads in wet deposition are not very well characterized. Also the contributions of gaseous ammonia, volatile organic acids and amines, and various nitrogen and sulfur aerosols to total nitrogen and sulfur loads are not very well characterized.

At the same time, despite some of these uncertainties about specific air concentration, total loads, and ecosystem responses, however, the general quantitative relationships between regional decreases and increases in total emissions of nitrogen and sulfur relationships and several different nutrient enrichment and acidification effects are reasonably well characterized.

9. To what extent has the draft ISA adequately characterized the contribution of oxidized and reduced forms of nitrogen to ecological effects related to nutrient enrichment?

The present Draft ISA does a pretty adequate job of characterizing the contribution of oxidized forms of nitrogen to nitrogen enrichment but has a long way to go with regard to the contribution of reduced forms of nitrogen to nutrient enrichment.

10. Several additional effects are discussed, including mercury methylation, direct gas-phase effects on foliage, and N₂O as a greenhouse gas. How well does the draft ISA characterize the evidence on these topics?

My impression is that the present draft ISA does a reasonably adequate job of describing the available evidence on these other topics. I gather from some of the Charge Questions in the Draft Scope and Methods paper, however, that EPA is still trying to make up its mind about how much attention to give to these other effects that go beyond the general issues of nutrient enrichment and acidification effects in both terrestrial and aquatic ecosystems.

11. What are the views of the Panel on the appropriateness and comprehensiveness of the conclusions drawn in Chapter 5?

I could not be more delighted with the decision to provide this kind of bulletized presentation of major Findings and Conclusions from the various chapters of this draft ISA! It appears that a considerable and well-focused effort has been made to develop this series of very carefully crafted brief summary statements that:

- 1) Contain the distilled essence of the most important topics covered in each chapter, and
- 2) Are as directly relevant as possible to the overarching Key Policy Questions that should be the principal focus of all aspects of these NAAQS review processes:.

“What scientific evidence and/or scientific insights have been developed since the last review to indicate if the current public-health based and/or the current public-welfare based NAAQS need to be revised or if alternative indicators, levels, statistical forms, or averaging times of these standards are needed to protect public health with an adequate margin of safety and to protect public welfare?”

“What scientific evidence and/or scientific insights have been developed since the last review to indicate whether, and if so, what particular ecosystem components or other air-quality-related public welfare values, are more or less sensitive than the populations of humans for which primary standards are established and for this reason may require a different indicator, level, statistical form, or averaging time of a secondary standard in order to protect public welfare.”

12. How adequate is the draft ISA for providing information and guidance to future exposure, risk and policy assessments that may be prepared in support of this NAAQS review?

I am a bit confused by this question. At first, I thought the question related to needs for information and guidance for future NAAQS reviews, but then the question ends with "policy assessments that may be prepared in support of this NAAQS review?"

In any event, I do not recall that very much attention has been given in this First Draft ISA to providing information and guidance with regard to either future or the present reviews for secondary NAAQS standards for nitrogen and/or sulfur.

Dr. Douglas Crawford-Brown

This review focuses primarily on Chapter 5: Findings and Conclusions, although it draws on information in the earlier chapters from which the Findings were drawn. My first very general comment is that the Findings did, I believe, comport with the major conclusions one might draw from the earlier chapters. At least, they summarized what I took to be most of the main points in these earlier chapters, although I caution that I am not an ecologist and so there may have been significant points made earlier that did not make their way into Chapter 5.

It was a bit hard (perhaps too hard) to get the subsections of Chapter 5 to match the way the earlier chapters were organized. This is due largely to the fact that Chapter 5 itself does not have any systematic approach to presenting the findings and conclusions concerning effects (which is really the focus of the NAAQS process). The effects are divided between Acidification; Nitrogen Nutrient Enrichment; and Other Welfare Effects. Then these three major classes of effects are subdivided in various ways (such as Aquatic; Terrestrial; etc). However, the subdivisions are not consistent across the classes of effect. I suppose this might be because some of the subdivisions are relevant for one class of effects and not others, but I can't see any reason why this should be the case. It would have been much more helpful to have the three major classes subdivided into the same set of subsections so the reader can quickly go through and find, for example, the Biochemical Effects for all three classes. The needed information is all there, I just wanted a more consistent structure to the presentation of it.

I also didn't understand the case studies. I could find no consistency in the way they were approached, or any conclusions that could be drawn from them. There was no explanation of what they are doing within the document (i.e. what the reader is to take from them). They struck me as an arbitrary set of rather poorly detailed cases – I'm sure the authors had some reasons for selecting them, but this was not evident.

My final general quibble is with the highly qualitative nature of many of the conclusions on ecological effects, and the general presumption that an effect is the same as an adverse effect. The chapter is littered with phrases such as “could cause”, “are thought to”, etc. There is an implicit assumption throughout that ecosystems are in their optimal states before NAAQS-relevant sources are added to the world, and that all these sources can do is weaken ecosystem health. Perhaps the authors don't intend this implicit assumption, or the impression that the phrases I mention count as strong evidence for any sort of belief, but I was left uneasy with many of the conclusions for the reasons above.

I now have some specific comments on parts of Chapter 5:

1. On Page 5-2 at the bottom, the authors conclude that measurements below the detection limit cause “irresolvable uncertainty in these data”. I agree with the sentiment here, but there are policy and regulatory decisions that can be made that don't have this “irresolvable uncertainty”. For example, if the regulatory limit is well above the detection limit, and all the positive results are below the regulatory limit, then a large number of results below the detection limit does not

prevent a decision as to whether a site is in compliance to be made with high levels of certainty. Whether the “irresolvable uncertainty” has any implications for decisions depends entirely on the kind of decision being taken.

2. On Page 5-4, first bullet, the authors state that “annual NO_x has decreased < 35% over the years”. It was not clear whether this is to mean it has decreased by less than 35% of the baseline year (1990) or has decreased TO LESS THAN 35% of the value in the baseline year.

3. On Page 5-6, the authors correctly state that it is possible that small amounts of SO₂ may be beneficial. It would be useful to indicate how this amount compares to the levels considered under a NAAQS and the levels currently found in the environment. If the amounts that are beneficial overlap the amounts found in the environment, this has significant implications for the NAAQS. And it also would be useful to characterize the evidence for beneficial effects, since it indicates that the exposure-response curve may be U-shaped. Focusing only on the “bad” effects leaves the document open to charges of bias and presumes there is some sort of monotonic increase of bad effects with exposure, or a threshold below which effects don’t occur (but also are not beneficial).

4. In that same paragraph, the authors state that lack of observation of a change doesn’t mean that no change is taking place (it may have been below the ability to detect such a change). I just want to be sure it is also understood that this statement cannot be used to therefore justify regulatory action; the evidence simply becomes neutral with respect to decisions being taken.

5. On Page 5-11, the second bullet, the authors conclude that a lack of relationship may indicate that the trends are on a time scale longer than is being measured. Well, yes, this could be the case. Or it could just be that there IS no relationship. I find this pattern throughout the document: a tendency to explain away a lack of relationship as being due to some limitation in the data, rather than the simpler, and more truthful, claim that there has been no trend observed to date.

6. On Page 5-12, first bullet, line 16, the term “likelihood” is used. I couldn’t figure out the sense in which it is being used here. I assume it is not in the statistical sense.

7. On Page 5-15, last full paragraph, the final sentence doesn’t seem to me to follow from the evidence presented, unless one assumes that ANC values are the sole determinant of acidic episodes. I won’t comment further on this, because it is not my area of expertise, but this implicit assumption must be made and so I was left wondering where the evidence was given to support it.

8. Throughout the discussion on NO_x and SO_x deposition, I could not find any recognition, or at least explicit consideration, of deposition onto land that then enters a waterbody through run-off. I am assuming it is being considered even if not called out directly in the text.(?)

9. On Page 5-17, last bullet, the issue of Hg increasing in fish is brought up. I realize that the authors are suggesting here that this increase above “safe” levels is due in part to the effects of acidic deposition, but this section is focused on effects and no specific effect is mentioned (unless one takes the increased Hg itself as an effect, which is what I presume the authors

intend). But there should at least be some mention of the extent to which this increase in Hg is due to the effects of acidic deposition and not just to loading of the original Hg into the waters.

10. On Page 5-18, first paragraph under 5.5, we find another instance in which a change is automatically considered adverse. The argument is that excess N creates “unnatural rates in some species and change in the competitive interactions...”. This is then followed by the claim that this will decrease ecosystem health and biodiversity. Again, I am not an ecologist, but my reading of the literature suggests it is far from evident that all such changes weaken ecosystem health (unless one assumes *a priori* that ecosystems are in their optimal state of health absent increased N). Or the key may be in the term “excess”. Perhaps the authors have some idea as to what level of N constitutes an “excess” and are referring only to this condition, in which case we have a tautology (the level of N that is “excess” being by definition the level of N that causes weakened ecosystem health). Without addressing this issue, the reader is likely to assume that “excess” is the same as “increased”, which would not be true.

11. On Page 5-19 at the top, it seems to me that there is an assumption being made here that all N in the system is bioavailable. I may be wrong in this, but it does seem to me that assumption is inherent in the paragraph. At the bottom of that same page, the authors refer to the “availability of NO₃-. I wonder if they mean “bioavailability” or if the two terms are interchangeable.

12. On Page 5-21, line 13, it is noted that a change in shoot-to-root ratio can be adverse. I would guess it is adverse primarily if the ratio increases, not decreases.

13. On Page 5-26, line 12, it is mentioned that the function is not linear, but no indication is given of the kind of non-linearity it represents. Some clarity here would be good (threshold, positive second derivative, negative second derivative, U-shaped, etc).

14. The Regional Trends sections struck me as unsatisfactory throughout the chapter. I was never clear, in any of them, what important conclusions, related to a NAAQS decision, I was to take away. It wasn't even made clear why a regional trend is of interest.

15. On Page 5-31, a list of biological measurements to assess eutrophic condition is provided. But I don't see why this list is here. I can find no way in which it informs the later discussion or conclusions.

16. The discussion of Greenhouse Gases beginning on Page 5-35 is confusing, or at least given in too much of a sketch. I haven't any idea what the reader is take away from this.

17. On Page 5-36, the first sentence states that in the chapter data were “integrated and collectively considered in formulating conclusions”. I don't think the chapter accomplished this, or made it evident. I am not saying the conclusions are wrong, or that the authors didn't have good reasons for giving them. I am simply saying that the chapter doesn't lay out all clearly how the data were “integrated and collectively considered”. I would be more inclined to say that the data were collected together, the authors considered them, and conclusions were formulated (in ways that are not transparent to the reader).

18. On Page 5-37, line 11, there is an instance of something that appears in other places in the chapter. Some N and S deposition rates are mentioned, but there is no indication given as to whether these are large or small compared to deposition rates one would expect under NAAQS levels. The reader is left, therefore, wondering whether these deposition rates are significant with respect to any decisions that must be taken under NAAQS.

19. The final sentence of the chapter is that “The Chesapeake Bay is an example of a large well-studied estuary that receives 21 – 30% of its total N load from the atmosphere”. This seems a very odd way to end the chapter, as this sentence is essentially a *non sequitur* and certainly doesn't summarize any of the important conclusions.

Dr. Charles T. Driscoll, Jr.

I have a few general comments that I will relay to you in this letter and the enclosed file has my detailed comments. I was disappointed in the ISA. The scope is generally fine but the text is not polished. It does not appear that the document was particularly well edited. As a result, I have considerable comments.

I have a few concerns with the document. Sections are highly redundant. With a bit of editing, I would guess that the length could be decreased 25%. The referencing of the document is uneven. There are many instances where facts are stated that are not common knowledge and no citation is provided. I would think that this approach would not be acceptable. Other sections of the document are referenced adequately.

There are several statements that are made which are incorrect. I have either noted these or corrected the text.

I do not like the approach as on 1-6, line 26 “Chapter 2 highlights”. This is grammatically incorrect. A chapter is an inanimate object and cannot highlight anything. This is done throughout the document. It should be corrected.

The section on sampling and analysis for NO_x and SO_x (2.6) while interesting, is long and could easily be put in an Appendix.

I understand that I really don't have much say in the matter, but interests would be best served if the scope of the ISA was expanded to include base cation and Cl^- deposition. Cl^- is probably largely derived from coal combustion or industrial processes at least away from the coast. Both base cations and Cl^- deposition can influence the acid-base chemistry of ecosystems.

- In Chapter 3 (3-28), shouldn't some mention be made of cloud deposition?
- In this section (3-28, 15), it is mentioned that NH_3 is not included in deposition estimates. Another problem that isn't mentioned is DON deposition. This should be added to the text.
- The document is made confusing by mixed units and symbols. Mass and molar, English and metric. It seems that there is no unit or scale that is not used in this document.
- Also different ways of expressing units are used (e.g., kg/ha-yr vs. $\text{kg ha}^{-1} \text{yr}^{-1}$). A consistent format should be used throughout the document.

**Specific Comments on Integrated Science Assessment
for Oxides of Nitrogen and Sulfur in Environmental Criteria**

Chapter 2

Page 2-2, line 5	Reference needed.
Page 2-3	Figure title: Define M, PAH, PAN
Page 2-4, line 6	Change to: ... (e.g. nitrosamines, nitro-PAHs)
Page 2-5, line 23	Change to: ... effectively no chemical or physical removal mechanism in the
Page 2-10, line 24	Make italics: <i>in situ</i>
Page 2-11, line 4	H ⁺
Page 2-11, line 12	Change to: ... compounds affect
Page 2-14, line 6	Change to: ... acidity, at adequate concentrations it affects...
Page 2-14, line 19	Change to: For any [NH _x] in the system... condensed phase (Reaction 2.4-4).
Page 2-16, line 17	Change to: ... Earth's surface...
Page 2-19, line 27	Change to: oversaturated with
Page 2-21, line 22	Make italics: <i>in situ</i>
Page 2-21, line 25	Change to: An overview of the three satellite... backscatter is contained in Table 2.6-1.
Page 2-21, line 27	Change to: Total column [NO ₂]... satellite is shown on Figure 2.6-1.
Page 2-23, line 9	Change to: ...; see the more complete description ...
Page 2-28, line 12	Change to: Excess NH ₃ alone or together with NO _x can enhance to terrestrial and aquatic ecosystems.
Page 2-29, line 7	Change to: ... networks is sparse over...

Chapter 3

Page 3-1, line 11	Change to: In this section brief summaries...(NH ₄ ⁺) are provided.
Page 3-2	Section 3.1.2.1 Soils. This section needs some citations.
Page 3-2, line 20	Isn't N ₂ the dominant soil gas?
Page 3-2, line 30	Is there a typo here? Do you mean "Although N ₂ O is not reactive...?"
Page 3-3, line 4	Do you mean...oxidation of NH ₃ released?
Page 3-3, line 24	Is there a word or something missing? This sentence doesn't make sense.
Page 3-3, line 28	What is meant by fuel N loadings? Clarify.
Page 3-3, line 31	What section?
Page 3-4, Line 3 & 4	Need a reference
Page 3-4, line 23	Change to: ... and this pattern remains...
Page 3-5, line 5	This sentence doesn't make sense, please clarify.
Page 3-5, line 9	What is meant by "stimulated through soil management"?
Page 3-5, line 12	You don't really want the brackets here do you?
Page 3-5, line 29	Cultivation of soil with high organics content is not an addition of N as indicated in the sentence that follows.
Page 3-6, line 1	Change to: ... of which a small portion
Page 3-6, line 30	Change to: Data for SO ₂ emissions...state level totals are depicted in Figure 3.2-1.
Page 3-7, line 6	Change to: The magnitude and spatial distribution... in the ARP is depicted in Figure 3.2-2 for the CONUS.
Page 3-7, line 7	Change to: continuing elevated density of SO ₂ emission sources in the ... compared to the West, particularly in the Central Ohio River Valley.
Page 3-9, line 2	Change to: ... increases with increasing fire intensity.

Page 3-9, line 12	Change to: ... emissions are lower. Reduced light levels...
Page 3-9, line 16	Change to: However, note that ...
Page 3-9, line 30	Need a citation
Page 3-12, line 23,24	Need citations
Page 3-14, line 7	Change to: The networks, sponsoring agencies... are listed in Annex Table AX2.5-1.
Page 3-20, line 1	Does CASTNET really have cation data? I never knew that.
Page 3-20, line 19	Change to: ... of ~ 90 sites. In addition the seven Atmospheric ... Also doesn't AIRMon provide gas and particle chemistry and estimates of dry deposition?
Page 3-22, line 31	Change to: Contributions from several... than 30 years are summarized in Annex Table AX2.5-5.
Page 3-23, line 9	Change to: Ambient [NO ₂] ... through 2005 is shown in Figure 3.7-1.
Page 3-24, Para. 1	Need citations
Page 3-25, line 6	Change to: ... evident along the Ohio ...
Page 3-25, line 7	Change to: ... have been decreasing throughout...
Page 3-25, line 9,10	Define CMSAs and LOD
Page 3-26	Figure title: Lower case c in concentration
Page 3-26, line 3	Change to: The composite diel... database is shown in Figure 3.7-4.
Page 3-27	Figure title: Clarify what is meant by "in focus".
Page 3-27, line 1	Change to: ... by sources some distance above the Earth's surface.
Page 3-33, line 3	Change to: Note, however, ...

Page 3-33, line 26	Change to: ... are problematic because
Page 3-34, line 4	Change to: ... a similar relative decrease
Page 3-35	Figure title: Clarify what is meant by O ₃ /10
Page 3-36, line 26	Change to: ... observed fluxes described here are compared in Figure 3.9-5.
Page 3-44, line 28	Change to: Results of a ..., respectively are shown in Figures 3.10-1 and 3.10-2.
Page 3-44	In the comparison of measured and modeled deposition (e.g. Figure 3.10-1) some discussion should be given on testing dry deposition fluxes. Also in Figure 3.10-1 the fluxes are given on an annual basis. What about patterns for other time intervals such as monthly or weekly? The scatter should be much greater. Should other predicted values for shorter time intervals be discussed?
Page 3-45, line 4	Why is this a critical load? This definition seems inconsistent with the remainder of the document. Please clarify.
Page 3-48	The statement in the first paragraph that atmospheric load is equal or exceeds riverine load (line 8) is inconsistent with the statement in the 4 th paragraph that atmospheric deposition is 10-40% of the total load. This needs to be clarified. Also a reference for the 10-40% contribution value needs to be referenced (Castro and Driscoll 2002?).
Page 3-50, line 4	Change to: Several waterbodies ... to total N loads are listed in Table 3.11-1.
Page 3-50, line 15	I do not believe the statement that 30-70% of the volume of animal waste is emitted as NH ₃ . This is not possible. The statement needs to be fixed.
Page 3-50, line 21	Change to: Several important watersheds ... airsheds are listed in Table 3.11-3 and 3.11-4.
Page 3-50, line 23	Reference is needed to justify the statement on larger airsheds for oxidized N.
Page 3-51, line 1	Change title: 3.12 Background (PBR) concentrations and deposition of NO _x and SO _x

Page 3-51, line 20	Change to: The annual mean [NO ₂] ...[NO ₂] (bottom panel) are shown in Figure 3.2-1.
Page 3-53	Give complete figure title. Don't be lazy.
Page 3-54, line 8	Change to: The highest values are ... Ohio River Valley (Figure 3.12-3 (upper panel)).
Page 3-54, line 23	Change to: Results from ... (Bey et al., 2001) is shown in Figure 3.12-5.
Page 3-55	Again, give complete figure title
Page 3-56	Again, give complete figure title
Page 3-59, line 7, 16	In addition to giving background concentrations, the summary should provide deposition values.
Page 3-59, line 12	Give the reader the specific percentage that EGUs contribute to SO ₂ emissions.
Page 3-59, line 29	Again, that statement about the NH ₃ contribution from animal waste is incorrect and needs to be fixed.
Page 3-60, line 21,26	Again, the inconsistent statement about the atmospheric contribution to the total estuarine input needs to be made consistent. The 10-40% value is the correct statement (Castro and Driscoll 2002).
Page 3-62	Is this the original reference? It is difficult to believe.

Chapter 4

** There needs to be a global search on this chapter on sulfate. It should be SO₄²⁻. The entire chapter needs to be fixed. Also throughout the document, the authors refer to “inorganic Al”. There needs to be a global search on this. This needs to be referred to as “dissolved inorganic Al” or “monomeric inorganic Al”. Much of particulate matter or soil is inorganic Al. The authors want to refer to dissolved inorganic Al. **

Page 4-1, line 8	Change to: A discussion of acidification is presented in Section 4.2. Nitrogen (N) enrichment is discussed in Section 4.3.
Page 4-1, line 19	Change to: ... , information is presented in this Integrated Science Assessment (ISA) that was collected...

Page 4-3, line 1	Change to: Critical loads as a ... quantifying disturbance is discussed in the following section.
Page 4-7	Paragraph 1: There is no discussion of target loads here. Should the text be expanded to address the concept of target loads?
Page 4-7, line 24	Change to: ... is the accumulation of hydrogen ion (H^+) ...
Page 4-7	Section 4.2.1.1: This section on soil acidification is woefully inadequate. Soil acidification is really the loss of base cations plus the accumulation of acidic anions. The authors should refer to van Breemen et al. (1983) or Binkley and Richter (1987).
Page 4-9	The incorrect figure is with this figure title.
Page 4-10, line 1	The statement of mobility of sulfate governing most aspects of soil and water acidification is simply not true and demonstrates a complete lack of understanding of the process. This needs to be corrected.
Page 4-10, line 6	Change to: ... deposited S is transported to the soil ...
Page 4-10, line 7	Change to: ... acts as a mobile anion at ...
Page 4-10	Paragraph 2: References are needed to document statements made in the paragraph.
Page 4-10, line 11	Change to: ... leaching of cations, and ...
Page 4-10, line 13	Change to: ... When S is transported from
Page 4-10	Last paragraph: You need to clarify why accumulated sulfate is slowly released from soil.
Page 4-11, line 2	Change to: ... the accumulation of the historic legacy of atmospheric S deposition in soil was ...
Page 4-11, line 9	Change to: ... and the difficulty in discerning the effects of net SO_4^{2-} description and net S mineralization make it difficult...
Page 4-12	Paragraph 1: Virtually all mass balance studies show N retention. See Campbell et al. (2004) for example.
Page 4-12	Paragraphs 1 and 2: References are needed to document the statements made.

Page 4-12, line 9	Change to: ... between atmospheric N deposition and the C:N...
Page 4-13, line 3	Add ...leaching to surface waters in the eastern United States (Aber et al. 2003)
Page 4-13, line 8	Change to: ... Although concentrations of NO_3^- are typically less than SO_4^{2-} in drainage waters...
Page 4-13, line 12	Change to: ... leaching from forest ecosystems...
Page 4-13	Paragraph 3 – References are needed...
Page 4-13, line 14	Clarify sentence
Page 4-13, line 20	Change to: ... most noteworthy effect of
Page 4-14, line 1	Change to: ... This pattern was likely due ...
Page 4-14	Paragraph 2: References needed
Page 4-14, line 21	Change to: ... S and N in acidic deposition enhance inputs of strong acid anions that can accelerate ...
Page 4-14, line 25	Change to: ... plant nutrients from soil; and...
Page 4-15	Paragraph 1: Reference needed....
Page 4-15, line 10	Change to: ... documented decreases in base saturation of ...
Page 4-15, line 16	This statement is not correct. Likens et al. (1996) and Kirchner and Lydersen (1995) documented decreases in stream concentrations of base cations due to decreased leaching from the soil exchange complex. They document decreases in soil exchangeable pools of base cations due to elevated acidic deposition.
Page 4-15, line 27	Should cite Cronan and Schofield (1990)
Page 4-15, line 29	Change to: ... deposition because inorganic monomeric ...
Page 4-16	Paragraph 2: References needed....
Page 4-16, line 30	Change to: ... deposition tends to remain in solution ...
Page 4-17, line 1	Need to clarify this sentence. Changes in concentrations of base cations do not necessarily result in increases in dissolved inorganic aluminum.

Page 4-17, line 8,9	Change to: ... as chronic condition or episodic condition. Chronic condition refers to annual ...
Page 4-17, line 11	Change to: ... Episodic condition refers to ...
Page 4-17, line 12	Change to: ... provide less neutralization of
Page 4-17, line 18	Change to: ... acidic deposition likely has substantially increased ...
Page 4-17, line 20	Change to: ... Many streams that exhibit chemical conditions during ...
Page 4-17, line 24	Need to define ANC
Page 4-17, line 25	Change to: in the central Appalachian Mountain region ... (a reference is also needed to document this statement)
Page 4-18	Figure title. The figure is incorrect for the figure title and text citation. Also <i>in situ</i> should be in italics.
Page 4-19, line 19	The statement here is not exactly correct. The difference between summer and spring ANC during baseflow conditions was on average 30 µeq/L. This means that acidic episodes would occur on average when summer ANC values reached 30 µeq/L.
Page 4-19, line 23	Define: episodic ANC
Page 4-19, line 28	Change to: ... contact with ANC supplying materials ...
Page 4-20, line 2	Change to: ... this pattern can be ...
Page 4-20, line 6	Reference needed ...
Page 4-21, line 1	Make italics: <i>in situ</i>
Page 4-21, line 4	Change to: ... during low flow, there is a shift to conditions of moderate to severe episodic acidification during high flow that showed higher ...
Page 4-22, line 28	Change to: ... 2002), larger-term trends in ...
Page 4-22, line 28,29	Sentence not clear, please clarify
Page 4-23, line 12	Should the title be Terrestrial Ecosystems or Forest Ecosystems?

- Page 4-23 4.2.2. Terrestrial Ecosystems. Should enhanced leaching of nutrient cations from the canopy due to acidic deposition be mentioned in this section?
- Page 4-24 Paragraph 1: Reference needed
- Page 4-24 Paragraph 2 and elsewhere: Throughout the document the authors discuss soil horizons as though they are always and only Spodosols. I don't believe that the only sensitive soils are Spodosols. Aren't other soil types important? When the authors refer to O and B horizons, they need to clarify the geographic context they are discussing, and if they are referring to all Spodosols.
- Page 4-24, line 10 Why is the Bs horizon more sensitive than the Oa horizon in the forest floor? I believe that the forest floor is more susceptible to cation change from acidic deposition than the mineral soil. Most of the roots are in the forest floor. Is a 20% loss of Ca^{2+} in the forest floor less important than a 50% loss of Ca^{2+} in the mineral soil?
- Page 4-25, line 2 Do we actually know that Al only becomes mobilized after Ca^{2+} becomes depleted? If this is true (I don't believe it) how about a reference documenting it?
- Page 4-26, line 10 Need a reference
- Page 4-26, line 17 Change to: ... nitrification rates are difficult to measure directly ...
- Page 4-28, line 2 Do you mean terrestrial or forest?
- Page 4-29, line 4 Has there been red spruce decline in the Southeast? Please provide a reference.
- Page 4-29, line 14 Need a reference
- Page 4-31, line 25,26 Change to: ... A conceptual view...maple decline is provided in Figure 4.2-2.
- Page 4-33, line 6,7 Clarify sentence
- Page 4-33 I think Juice et al. (2006) shows some compelling data on sugar maple response to Ca addition. The authors should consider mentioning this work.

Page 4-34, line 2,3	This section is on tree health. Why mention soil cation depletion here? Wouldn't it be more appropriate in the soil effects section?
Page 4-40	Note that while temporal patterns in surface water NO_3^- loss are confusing and variable. Aber et al. (2003) shows that there are spatial patterns across the region which are consistent with atmospheric N deposition contributing to elevated NO_3^- leaching.
Page 4-41, line 18	Change to: ... Efforts to explain the complex patterns in ...
Page 4-43, 14-16	Likens et al. (1998) showed increased losses of Ca^{2+} with flow during the 1960s and a declining pattern in later years.
Page 4-44, line 18	How do Shenandoah streams show changes in soil sulfate adsorption? Please clarify. One important observation that is ignored in this report is that although surface water ANC appears to be increasing, soil losses of exchangeable cations are continuing despite reductions in acidic deposition. This observation should be mentioned somewhere in the document.
Page 4-46, line 8-10	How large is large?
Page 4-46	Do you really believe this statement is true? Virtually all studies that measure major ion chemistry measure pH. I believe that ANC is more commonly focused on because it is more straight forward. pH is a non-linear measurement and its changes are difficult to interpret as trends.
Page 4-47	The statement that waters with $\text{ANC} < 0 \mu\text{eq/L}$ have "no capacity to neutralize acid inputs" is not true. An ANC value below $0 \mu\text{eq/L}$ simply means that H^+ values are elevated (the solution pH is below the equivalence point). This statement needs to be corrected.
Page 4-48, line 7,8	Change to: acidification (a decrease in ANC observed...)
Page 4-48, line 15	Where is Table 4.2-4?
Page 4-49, line 1	The units should be $\mu\text{eq/L-yr}$.
Page 4-49, line 2	Change to: ... represent significant trends towards...
Page 4-49, line 11	Change to: ... ANC below zero $\mu\text{eq/L}$.
Page 4-49, line 20	Add Driscoll et al. (1988)

Page 4-51, line 9,10	These conditions can also result in direct mortality as results from <i>in situ</i> bioassays show (Van Sickle et al. 1996).
Page 4-51, line 12	Add reference
Page 4-51, line 15	Change to: ... vigor, and reproductive success; and
Page 4-57, line 8	Table 4.2-6 – is missing
Page 4-57, line 15	Change to: ... relationships are complex, however, ...
Page 4-57, line 22	Change to: ... However, note that effects...
Page 4-60, line 16	Change to: Note that the absence ...
Page 4-61	Figure: It would be helpful if a comment was made on the consistency of Figure 4.2-11 with 4.2-10. Although the line 4.2-10 is different. The values on the two figures actually seem quite similar. Is fish diversity response to acidification similar in the Adirondacks and Virginia?
Page 4-66, line 6	What does this mean evaporative concentration? Please clarify.
Page 4-69, line 1	Change to: ... conditions in the Park for ...
Page 4-69, line 14	Elsewhere in the document Al concentrations are expressed in $\mu\text{mol/L}$.
4-70, line 22	Aren't the USDA Forest Inventory Data (FIA) available? Please clarify.
Page 4-71, line 9	Change to: Note that the McNulty...
Page 4-72, line 1	Red spruce are generally found at higher elevations.
Page 4-72, line 5	Change to: ... sugar maple which are
Page 4-72, line 7	Change to: ... areas where sugar maples appear to be ...
Page 4-73, line 28	Change to: Note that critical loads...
Page 4-74	4.2.4.3.2: The text in this section is redundant with the previous material presented.
Page 4-74, line 20	Give the specific %.

Page 4-76, line 11	Change to: ... 1991 levels of stream area that was suitable for brook trout survival
Page 4-76, line 18	Change to: ... required to restore watershed soils... (Note throughout the document, the authors use the term buffering capacity incorrectly. Please correct this.)
Page 4-76, line 25	Change to: ... was partly because SO_4^{2-} adsorption Also throughout the document, the authors refer to sulfur adsorption. I believe it is more appropriate to refer to sulfate adsorption.
Page 4-77, line 30	Change to: ... For aquatic ecosystems,
Page 4-79, line 30,31	Change to: ... ANC have been increased through liming...
Page 4-80, line 14	Change to: ... (e.g., pH, Al, Ca, ANC, DOC, dissolved OC)... Why do the authors use the term dissolved OC? Why not use DOC? Most readers will be familiar with this term.
Page 4-81, line 4	Change to: ... increased with lake pH and
Page 4-81, line 20	Change to: ...low weathering rates (Driscoll et al. 1991, Sullivan et al. 2006a)
Page 4-82, line 2,3	Need a reference
Page 4-82, line 25,26	Need a reference
Page 4-84	Figure 4.2-14: Why not update this figure and show the most complete time series of precipitation chemistry?
Page 4-84, line 23	Change to: ... The hydrogen ion deposited
Page 4-87, line 6	Change to: ... model simulations coupled with population-level extrapolations suggest that ...
Page 4-87, line 23	Change to: ... cations, hydrogen ion and
Page 4-89, line 15	This statement is not exactly true and needs to be qualified. These authors provided an upper limit of organic acid content and as a result greatly understated the number of fishless lakes due to acidic deposition. This is a very important mistake and needs to be clarified.

Page 4-89, line 22	Change to: ... pH and ANC decreased substantially...
Page 4-89, line 26	Note that Van Sickle et al. (1996) was part of the ERP and not subsequent to it.
Page 4-90, line 14	Change to: ... from 1982-2000 in the original 16 ALTM ...
Page 4-90, line 15,16	Change to: ... from 1992 to 2000 in the complete set of 48 ALTM lakes. (Note that 32 additional lakes were added to the program in 1992). They found ...
Page 4-92	Figure 4.2-18. You have the incorrect citation for this figure. It should be Driscoll et al. (2003a).
Page 4-92, line 4	The population-base estimates need to be mentioned earlier in the text where this study is first mentioned.
Page 4-93	Figure title: Change to: ... ANC at three dates for the population.
Page 4-94, line7	Note this citation is not correct. Chen and Driscoll (2004) applied the model to the DDRP lakes.
Page 4-94	Paragraph 2: Why not add a sentence or two on the modeled changes in zooplankton and fish species diversity that are discussed in Sullivan et al. (2006b)?
Page 4-94, line 30-32	No! Watersheds that are sensitive to mercury deposition are forested, have an abundance of wetlands, shallow hydrologic flow paths, are unproductive and impacted by acidic deposition (Driscoll et al. 2007).
Page 4-95, line 27	Change to: ... streams in the Park is linked ...
Page 4-95, line 28	Change to: ... soils to adsorb SO_4^{2-} is decreasing due to the long-term accumulation of SO_4^{2-} on soil adsorption sites associated with a legacy of elevated acidic deposition.
Page 4-97, line 14	Change to: ... At the low-ANC ($\sim 0 \mu\text{eq/L}$) ...
Page 4-97, line 16	Change to: ... Increases in base cations tended to compensate...
Page 4-97, line 26	Change to: ... NO_3^- concentrations usually increased...
Page 4-97, line 29	Change to: ... to the changes in ANC of Paine Run...River, and contributed to decreases in ANC in Piney River.

Page 4-98, line 3,4	Note that base flow ANC is controlled by bedrock geology.
Page 4-98, line 6	Note during high flow, the shallow flow paths diminishes contact between ...
Page 4-98, line 5	Change to: ..., probably because SO_4^{2-} adsorption ...
Page 4-99, line 1	Change to: ... more vulnerable to adverse effects of episodic acidification ...
Page 4-101	Paragraph 1: Isn't this material redundant with p. 76?
Page 4-101, line 8,9	These categories should have been defined previously in the text when they are first mentioned.
Page 4-101, line 31	What are sensitive Si-based watersheds? Please clarify. Somewhere the definition of sensitive southeastern watersheds needs to be defined: siliciclastic, granitic, and basaltic
Page 4-101, line 32	Is the loading kg S/ha-yr or as SO_4^{2-} ? Clarify.
Page 4-102, line 1	Change to: ... Prior to the Industrial Revolution, most
Page 4-102, line 3	Again, define Southeast watershed sensitivity and be consistent using it throughout the text.
Page 4-102, line 10	Clarify what is meant by small areas
Page 4-104, line 7,8	Here you finally define the watershed sensitivity classes. Use these terms throughout the text.
Page 4-104, line 25	Change to: ... N is transported between air ...
Page 4-104, line 27	Change to: ... because it is transported from the
Page 4-104, line 31	Change to: ... Leaching from soil,
Page 4-104, line 32	I don't believe that nitrate leaching from atmospheric deposition results in a violation of drinking water standards (>10 mg N/L). Where? I defy you to provide a reference(s).
Page 4-105	Also mention causes of declines in submerged aquatic vegetation (SAV) and causes of increases in nuisance algae species.
Page 4-105, line 3,4	see P. 112

Page 4-105	Note that nitrogen is also part of proteins, not only enzymes.
Page 4-106, line 29	Change to: ... N leaches from soils to
Page 4-109, line 21,24	Delete parenthesis from NO_3^- plus NH_4^+ on both lines
Page 4-111, line 5-7	Redundant. Denitrification has been defined previously.
Page 4-112, line 11,12	See below inconsistent with plot on p. 105
Page 4-112, line 19	Change to: ... are generally thought to reduce ...
Page 4-112, line 22,23	The methane response to nitrogen addition is confusing and inconsistent. This section should be rewritten.
Page 4-113, line 5	Change to: Note that the N enrichment...
Page 4-114, line 2	Change to: ... in the eastern United States (Driscoll et al. 2003a).
Page 4-114, line 21	Change to: ... fresh surface waters are: (1) elevated...
Page 4-114, line 22	Change to: ... water; and (2)
Page 4-114, line 24,25	Need reference
Page 4-114, line 26	Change to: ... in the Adirondacks (Aber et al. 2003)
Page 4-116, line 22	Change to: ... This pattern suggests that N ...
Page 4-116, line	Should you mention increases in populations of nuisance algae?
Page 4-119, line 15	Change to: ... not generally account for all wastewater inputs
Page 4-119, line 16-23	Should also cite Castro and Driscoll (2002).
Page 4-120	Change to: ... Source: (Driscoll et al. 2003b).
Page 4-126, line 15	These units do not make sense. What is the areal basis of the application?
Page 4-129, line 16	4.3.3.1.4 Herbaceous Plants and Shrubs – Move to forest section
Page 4-132, line 3	Change to: ... This pattern suggests selective ...
Page 4-132, line 14	Change to: ... In the reference plots, five species...

Page 4-132, line 14-16	I do not understand this sentence. Please clarify.
Page 4-135, line 1	This would be much more useful if expressed as a loading rate. The units shown here do not make sense mg/L-yr?
Page 4-135, line 32	Make italics: <i>in situ</i>
Page 4-138, line 17, 18	Make italics: <i>in situ</i>
Page 4-145, line 12	Make italics: <i>in situ</i>
Page 4-145, line 18	Change to: ... (e.g., cyanobacteria, dinoflagellates)
Page 4-146, line 1	Note Driscoll et al. (2003) is not the original citation. It should be (Valiela et al. 1990).
Page 4-147, line 27,28	Change to: An overview of the sensitive ecosystems is given in Table 4.3-2.
Page 4-152, line 9	Make italics: <i>in situ</i>
Page 4-152, line 28	Change to: ... future outlook of the U.S. estuaries based ...
Page 4-154, line 2	Change to: ... limited, though note that many
Page 4-160, line 26	Define very low. Isn't it generally defined as <2 mg/L?
Page 4-163, line 27	Change to: ... oxygen content, supply of labile organic carbon, temperature, pH ...
Page 4-165, line 2	Please clarify the mass basis of the loading Kg S/ha or Kg SO ₄ /ha?
Page 4-166, line 3,4	This statement is incorrect and misleading. High production of sulfide will limit methyl mercury production (Benoit et al. 2003). This sentence needs to be rewritten.
Page 4-166, line 30	Change to: ... watershed soils and the transport of naturally...
Page 4-168	Section 4.4.1.2.2: There is no reference to effects of mercury on song birds and terrestrial food chains (see (Rimmer et al. 2005).
Page 4-169	No reference is made of Drevnick et al. (2007) which suggests a link between declines in atmospheric S deposition and fish Hg.

Page 4-170, line 28	What is SO_4^{2-} deficit? I never heard of such a thing. Please clarify.
Page 4-171, line 26	Change to: ... Pollutants must be transported from the
Page 4-171, line 27	Change to: ... Although the transport of pollutants...
Page 4-172, line 23-25	Change to: ... concentrations of SO_2 . The effect of SO_2 on vegetation are summarized including some discussion of the limited recent literature.
Page 4-176, line 27,28	Change to: ... In Annex 7, a short list of GHGs and the environmental factors.... of climate change are provided in Table 7-1. A comprehensive...
Page 4-177, line 22	Change to: ... The additional laboratory study ...
Page 4-177, line 25	Change to: ... 145 yr old beech tree ...
Page 4-179, line 4	Change to: ... temperature, precipitation, and forest soil ...
Page 4-179, line 10	Change to: ... show increasing temperature increases ...
Page 4-179, line 21	Change to: ... ground water NO_3^- concentration is elevated ...
Page 4-179	Again, I can't believe that atmospheric N deposition results in surface water concentrations that approach drinking water standards. Please provide a reference.

Chapter 5

Page 5-3, line 11	Change to: ... networks is greatly limited over large ...
Page 5-5, line 23,24	As indicated before, this statement is not correct and inconsistent with lines 25 and 30 immediately below. Please fix.
Page 5-6, line 1	What ecological effects does S deposition have on Chesapeake Bay?
Page 5-6, line 14	Change to: ... including topography vegetation, soil chemistry ...
Page 5-8, line 9	Change to: ... There are several indicators of stress...
Page 5-9	Paragraph 1: Need reference.

Page 5-9	Bullet: A number of studies show ongoing soil acidification (i.e., net Ca ²⁺ loss) despite decreases in acidic deposition (Bailey et al. 1996, Likens et al. 1998, Huntington et al. 2000).
Page 5-9, line 24	Change to: ... as the pH decreases below 6.0
Page 5-10, line 14	Change to: ... which tend to provide less neutralizing of ...
Page 5-12, line 12	The sentence as written does not make sense. Please change. Change to: ... Decreases in pH below values of 6.0 typically
Page 5-14, line 19	Change to: ... in the pH range 5.0 to 6.0
Page 5-14	Fourth bullet – Change to: ... decrease in ANC below ____ µeq/L. Again, this sentence needs to be clarified. There is some threshold ANC below which effects are evident. My guess is that it is about 100 µeq/L.
Page 5-15, line 20	Change to: ... that where chronically acidic during summer in the ...
Page 5-16	Last bullet: The authors should clarify that stream surveys were not conducted in the Adirondacks or New England.
Page 5-16, line 28	Delete space before Maximum past ...
Page 5-17	Fourth bullet: Again, this is a best case scenario because the study overstated the level of acidity associated with organic acids. This needs to be clarified.
Page 5-17, line 30	Change to: ... with siliciclastic geology and ...
Page 5-19, line 19	Change to: ... elevation, climate, species, composition ...
Page 5-19, line 26,27, 28	Change to: : communities. The ecological effects...studied in recent years are summarized in Table 4.3-1.
Page 5-19, line 32	Change to: ... ecosystems. Note that N saturation...
Page 5-20, line 16	Delete first sentence. Start paragraph with ... In general forest ...
Page 5-23, line 1	Change to: ... More than 30 kg N/ha-yr of ...
Page 5-28	Last bullet. N fluxes ... to the total N budgets are compared in Table 5.5-1.

- Page 5-28, line 10 Change to: ... N to total phosphorus was ...
(Note, also clarify if this ratio is on a molar or mass basis.)
- Page 5-31, line 1 Change to: ... Si:N ratio decreases below....
- Page 5-31, line 6,7 Change to: ... in community composition, reduces hypolimnetic
DO, decreases biodiversity, and causes declines in submerged ...
- Page 5-32, line 26 Need a reference. Should the units here be metric?
- Page 5-33, line 30 Change to: ... influenced by oxygen content, supply of labile
organic carbon, temperature ...
- Page 5-36, line 11 Change to: ... average of 0.053 ppm. (space after 0.053)
- Page 5-36, line 19 Change to: ... Note that the regulatory....
- Page 5-37, line 13 Change to: ... forms of reactive nitrogen loading...
- Page 5-38, line 2 Change to: ... hypoxic zones, loss of habitat and harmful ...
- Page 5-39 Third section. Base Cations. Add references (Bailey et al. 1996,
Huntington et al. 2000).

References:

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Dr. Paul J. Hanson

My comments include general thoughts on the organization and content of the ISA, comments on specific chapters and items within the text, and summary thoughts on the charge question to which I was assigned.

General Comments:

The Integrated Science Assessment (ISA) provides a concise, but occasionally cursory overview of the key scientific issues for nitrogen and sulfur oxides related to atmospheric chemistry and physics (Section 2), ecological exposures (Section 3), and effects (Section 4). The section on effects is missing a brief discussion of the importance of NO_x as an ozone precursor. NO_x-induced tropospheric ozone effects need not be covered in detail since they have recently been reviewed as a part of the recent CASAC evaluation of photochemical oxidants. Nevertheless, the key role that NO_x plays as a precursor to ozone in the troposphere must be highlighted in the ISA.

The ISA would benefit from the addition of a section on the complexity of N and S biogeochemical cycles and the need to understand all natural and anthropogenic inputs and outputs to these cycles.

The most readable and best prepared section of the ISA was Section 5 on Findings and Conclusions. In fact, I recommend that Section 5 be brought forward (at least in part) to be presented as a summary of the documents key conclusions. Section 5 strikes the right balance between the beneficial and adverse effects of N deposition that was largely missing within Section 4. In a lead-off position within the ISA, the Findings and Summary material would provide the key conclusions from which an interested reader might then search for additional details and support within the document and its Annexes. All key conclusions within the Findings and Summary section must include parenthetical references to the pages within the ISA or appropriate Annex that the reader could look to find justifications for the conclusions.

For continuity with the previous Air Quality Criteria Documents (AQCD) for nitrogen and sulfur oxides, summary materials from those documents that have not been changed by new research might be brought forward and used within the ISA. For example, little new information has become available on the direct effects of NO_x and SO_x on plant response, but the quantitative understanding of the generally high ambient concentrations needed to illicit adverse responses to direct NO_x or SO_x exposures should be reiterated within the ISA. Section 5 provides the general statements of limited direct effects at current ambient concentrations, but Section 4 should include an overview of the key data from previous AQCDs in support of those statements.

When adverse effects are discussed within Section 4 of the ISA they should (where possible) be referenced to the state of current exposures presented within Section 3. For example, the authors need to show the reader how to interpret a multi-year experimental exposure to 20 to >100 kg N ha⁻¹y⁻¹ in the context of ambient levels that typically are maxed out near 10 kg N ha⁻¹y⁻¹.

The authors have not taken full advantage of the Annexes. By my count Annexes 1 and 2 were cited 7 and 11 times, respectively, but Annexes 3 through 10 were cited only 2,1,1,0, 1,0,0, and 1 times, respectively. The Annex material should serve as a source of expanded information

for the reader to highlight key points made within the ISA. In their current form and level-of-use the Annexes are not helpful to the reader.

References to primary research articles and cross-references to key discussions within the ISA and Annexes are often left out or ignored. This deficiency must be corrected. I offer some suggestions in the specific comments listed below.

A limited literature search for nitrogen deposition and impacts research conducted from 1991 through 2008 produced a number of research articles that are not mentioned or evaluated within the ISA.

As much as possible the authors need to limit the use of subjective statements like ‘maybe’, ‘if’, ‘probably’, ‘possibly....etc’. Rewriting such statements to indicate the true quantitative nature of the primary research conclusions would be a better approach.

Specific Comments:

Chapter 1

Page 1-2 lines 5 to 22: Add a bullet asking if the form of the current secondary standard is appropriate for the evaluation of and protection against adverse effects.

Page 1-5 lines 12 to 15: Even though no substantive research has been generated on the topic of gas phase responses to NO_x and SO_x since the publication of their most recent AQCDs, respectively, the ISA should include a brief overview and quantification of the air concentrations of gaseous forms of N and S necessary to generate adverse responses.

Chapter 2

Page 2-16 line 10: The term sensitive ecosystem is used here prior to it being defined in the context of the ISA.

Chapter 3

Page 3-11: Section 3.5.1 is inadequately referenced. The authors should either add references or point to a more detailed discussion within an Annex or to pages within a previous AQCD where such a discussion can be found. The following references might be quoted as a demonstration of how HNO₃ is incorporated into foliage:

- Hanson PJ, Garten CT (1992) Deposition of H¹⁵NO₃ vapor to white oak, red maple and loblolly-pine foliage -experimental-observations and a generalized-model. *New Phytologist* 122:329-337.
- Garten CT, Schwab AB, Shirshac TL (1998) Foliar retention of N-15 tracers: implications for net canopy exchange in low-and high-elevation forest ecosystems. *FOREST ECOLOGY AND MANAGEMENT* 103: 211-216.
- Vose JM, Swank WT (1990) Preliminary estimates of foliar absorption of N-15 labeled nitric-acid vapor (HNO₃) by mature eastern white-pine (*Pinus-strobus*) *CANADIAN JOURNAL OF FOREST RESEARCH* 20:857-860.

Page 3-11: A number of useful references could have been added/cited within this section. For example:

- Ammann M, Siegwolf R, Pichlmayer F, et al. (1999) Estimating the uptake of traffic-derived NO₂ from N-15 abundance in Norway spruce needles. *Oecologia* 118: 124-131.
- Boyce RL, Friedland AJ, Chamberlain CP, et al. (1996) Direct canopy nitrogen uptake from N-15-labeled wet deposition by mature red spruce. *CANADIAN JOURNAL OF FOREST RESEARCH* 26: 1539-1547
- Nussbaum S, Vonballmoos P, Gfeller H, et al. (1993) Incorporation of atmospheric (NO₂)-N-15-nitrogen into free amino-acids by Norway spruce *picea-abies* (l) karst. *Oecologia* 94:408-414.

- Qiao Z, Murray F (1998) Improvement of the N-15 dilution method for estimation of absorption of NO_x by plants supplied with N-15-labelled fertilizer. *New Phytologist* 138:13-18.
- Segschneider HJ, Hutzen H, Forstel H, et al. (1993) Uptake of (NO₂)-N-15 and metabolic transfer of the (NO₂)-N-15 nitrogen to various nitrogen fractions of sunflowers. *ISOTOPENPRAXIS* 29:51-57.
- Vallano DM, Sparks JP (2008) Quantifying foliar uptake of gaseous nitrogen dioxide using enriched foliar delta N-15 values. *New Phytologist* 177: 946-???
- Vonballmoos P, Nussbaum S, Brunold C (1993) The relationship of nitrate reductase-activity to uptake and assimilation of atmospheric (NO₂)-N-15-nitrogen in needles of norway spruce (*Picea-abies* [L] Karst). *Isotopenpraxis* 29: 59-70.

Page 3-11 lines 24 to 25: A reference is needed for this statement. Perhaps the following:

Hanson PJ, Rott K, Taylor GE, et al. (1989) NO₂ deposition to elements representative of a forest landscape. *Atmospheric Environment* 23:1783-1794.

Page 3-12 line 21: A reference is needed for the 1 ppb compensation point quoted here.

Figure 3.6-1: Why is this figure used? Shouldn't it be a summary of NO_x and SO_x issues?

Figure 3.6-2. The caption should read 'Aggregate map of the majority of routine U.S. monitoring stations.'

The Figure and brief paragraphs on page 3-17 seem unnecessary.

Section 3.7.1 would be improved with the inclusion of the graph demonstrating the nature of NO_x concentrations through time (i.e., over decades). Is this data not available? The graphics provided for SO_x and nitrogen and sulfur deposition in subsequent sections nicely demonstrate the declining nature of N and S inputs to most regions from the late 1980s early 1990s in when compared to 2004 to 2006 data.

For places where a complete continuous record exists it would be appropriate to include a graphic showing the full trajectory of N and S deposition through measured time.

Captions for the Figure 3.8-x series need to include a description of the source of the data within the figures.

Statements made at the top of page 3-41 need to be referenced to their source literature. In general, Section 3 suffers from inadequate attribution of the primary source data.

The total and background axes with the figures on pages 3-52, 3-53, 3-55, and 356 should be made the same. As drawn, the figures make it difficult to judge the level of background N or S levels against current totals. I would also recommend that the bottom graph in each of these figures be changed from a percentage of background to a quantitative amount of the indicated exposure measure above background. Low background levels near zero make the calculation of percent changes of little value. That is, a large percentage of a really small value is often still a really small value.

Page 3-58 lines 28: This is a key point. Where are the data or references to back it up? A citation to a publication, or another EPA report needs to be included here. Additional references are also needed on page 3-59.

Please consider bringing the following Annex figures forward to this section of the ISA: AX4.1-1, AX4.1-2, AX4.1-3.

Chapter 4

I would prefer to see this section organized along the lines of direct versus indirect effects of NO_x and SO_x. That approach has served the community well in the preceding AQCDs, and it better reflects that nature of the current N and S standards (gaseous exposures). Specifically Section 4.4.2 should be moved ahead of the discussion of acidification and nitrogen nutrient additions. This recommendation is not offered to perpetuate the use of a gaseous form of the welfare standard, but rather to recognize the history of NO_x and SO_x regulations. Delegating the primary effects of NO_x and SO_x exposures to a hidden corner at the back of the report seems inappropriate.

Page 4-2 line 1: The term biosynthesis may need further definition.

Section 4.1.3: The discussion of critical loads might be better located following the discussion of all direct and indirect effects. Redundant sections could all be combined together into a single section on the critical loads relevant to the combined and interacting effects of the full range of adverse effects of N and S inputs. Such a section should describe the need for a comprehensive biogeochemical cycling-based assessment of the N and S cycles in support of the concept of critical loads.

Page 4-6 lines 10 and 11: Where are the dose-response relationships?

Page 4-7 line 28: A reference is needed for this statement.

Page 4-8 line 1: A reference is needed for this statement.

Figure 4.2-1 contains the wrong image. It has been inadvertently swapped with the figure for the image in Figure 4.2-2.

Top of page 4-12: References to these statements are needed.

Page 4-12 lines 20 and 21: A reference is needed for the C:N statement.

Page 4-12 line 28: N may also be retained for long periods within plant biomass.

Page 4-12 line 29: A reference is needed for this statement.

At some point within Section 4 it would be useful to summarize the percent of US land area (perhaps within regions) anticipated to be impacted by direct effects of N and S forms, acidification, N deposition, etc. A table would suit this purpose nicely.

References are needed in Section 4.2.1.4 on base cation leaching.

Page 4-26 line 22: OC needs to be defined.

Section 4.2.2.2 is a good discussion (needing references) that might serve the reader better if it occurred earlier in the document.

Page 4-28 line 25: Would it be appropriate to change the word “gradual” to ‘accelerated’?

Page 4-29 lines 14 to 22: References for these statements are needed.

Page 4-31 line 10 and 11: The authors should be careful when using a phrase like “contributed to”. In many cases the data probably showed only a correlation with an observed effect rather than proof of a specific mechanism responsible for the effect. Similarly, on Page 4-31 line 26, should or could the word “indicated” be replaced with ‘hypothesized’?

Page 4-33 lines 19 and 20: Can this statement be made more quantitative?

Section 4.2.4.1 is an excellent section that is well referenced. Use it as an example when modifying other portions of Section 4.

Page 4-64 line 23: An “in review” paper is not an acceptable inclusion in the ISA.

Page 4-65 line 8: Were discussed regions acid naturally or have they developed acidity as a result of anthropogenic N and S additions? Can such a distinction be made?

Page 4-66 lines 26 to 28: Add the literature citation for this statement.

Page 4-70 line 21: I think this heading should be numbered. Perhaps 4.2.4.2.2.

Bottom of page 4-70 and top of page 4-71. The text should be expanded to show the components of the analysis of McNulty et al. (2007). The reader doesn’t have sufficient information. Is such information present in an Annex? If so, please direct the reader to the text. How are variable rates of plant productivity used in this analysis?

Page 4-71 line 30: A reference for this statement is needed.

Page 4-104 lines 14 to 24: This material is unnecessarily redundant with material in Sections 2 and 3.

Page 4-121 line 13: Is a change in productivity or biodiversity necessarily an adverse effect? This question should be discussed in the context of N and S deposition or loading.

Within section 4.4, the discussion on direct phytotoxic effects should be moved to the front of Section 4. The discussion of direct phytotoxic effects should include a description of the key exposure response relationships known to drive adverse direct effects. Such information could easily be lifted and reused from prior AQCDs since little new information is available on this topic. The following references published since the last N and S AQCDs and should be considered for inclusion in the ISA discussion:

- Ammann M, vonBallmoos P, Stalder M, et al. (1995) Uptake and assimilation of atmospheric NO₂-N by spruce needles (*Picea abies*): A field study. *WATER AIR AND SOIL POLLUTION* 85:1497-1502.
- Desantis F, Allegrini I (1992) Heterogeneous reactions of SO₂ and NO₂ on carbonaceous surfaces. *ATMOSPHERIC ENVIRONMENT PART A-GENERAL* 26:3061-3064.
- GunthardtGoerg MS, Schmutz P, Matyssek R, et al. (1996) Leaf and stem structure of poplar (*Populus x euramericana*) as influenced by O₃, NO₂, their combination, and different soil N supplies. *CANADIAN JOURNAL OF FOREST RESEARCH* 26:649-657.
- Kainulainen P, Holopainen JK, Oksanen J (1995) Effects of SO₂ on the concentrations of carbohydrates and secondary compounds in Scots pine (*Pinus-sylvestris* L) and Norway spruce (*Picea-abies* (L) Karst) seedlings. *New Phytologist* 130: 231-238.
- Manninen S, Huttunen S (2000) Response of needle sulphur and nitrogen concentrations of Scots pine versus Norway spruce to SO₂ and NO₂. *ENVIRONMENTAL POLLUTION* 107:421-436.
- MENG FR, COX RM, ARP PA (1994) Fumigating mature spruce branches with SO₂ effects on net photosynthesis and stomatal conductance. *CANADIAN JOURNAL OF FOREST RESEARCH* 24:1464-1471.
- Qiao Z, Murray F (1997) The effects of root nitrogen supplies on the absorption of atmospheric NO₂ by soybean leaves. *NEW PHYTOLOGIST* 136: 239-243.
- Qiao Z, Murray F (1998) The effects of NO₂ on the uptake and assimilation of nitrate by soybean plants. *ENVIRONMENTAL AND EXPERIMENTAL BOTANY* 39:33-40.
- Thoene B, Schroder P, Papen H, et al. (1991) Absorption of atmospheric NO₂ by spruce (*Picea-abies* L Karst) trees .1. NO₂ influx and its correlation with nitrate reduction. *New Phytologist* 117:575-585.
- Vassilakos C, Katsanos NA, Niotis A (1992) Physicochemical damage parameters for the action of SO₂ and NO₂ on single pieces of marble. *Atmospheric Environment* 26: 219-223.
- Wolfenden J, Pearson M, Francis BJ (1991) effects of over-winter fumigation with sulfur and nitrogen dioxides on biochemical parameters and spring growth in red spruce (*Picea-rubens* Sarg) *Plant Cell and Environment* 14:35-45.
- Wulff A, Karenlampi L (1996) Effects of long-term open-air exposure to fluoride, nitrogen compounds and SO₂ on visible symptoms, pollutant accumulation and ultrastructure of Scots pine and Norway spruce seedlings. *Trees* 10:157-171.

A brief section on the role of NO_x as an ozone precursor should be added to Section 4.4. This need not be very long, but it should provide sufficient information to inform the reader that NO_x pollution is often indirectly responsible for known adverse effects of photochemical oxidants on vegetation (and materials too for that matter). The reader could simply be pointed to the recent AQCD for ozone and other photochemical oxidants.

Some mention should be made of the potential for anthropogenic N additions to fertilize natural ecosystems leading to the sequestration of CO₂ from the atmosphere. This could be added as a counter point to the discussion of N₂O emissions to be expected from N saturated systems. If the authors don't provide this discussion and show that the issue was considered others will demand that it be added later.

The Tables attached at the back of Section 4 should be embedded with the text to which they apply or moved to their corresponding Annex.

Chapter 5

I really enjoyed reading Section 5. I found it to be concise, informative and easy to follow. As I've already stated I believe it (or a portion of it) could be presented at the front of the document. It does, however, need to be populated with references to appropriate portions of the ISA, ISA-Annexes or other published work that support each statement.

Page 5-2 line 13: Should "nitrification" be 'eutrophication'?

Page 5-4 lines 16 to 18: Although this statement may be true, I don't believe it is supported within the current draft of the ISA.

Section 5 might be rearranged to include:

5.4 Direct Phototoxic Effects of NO_x and SO_x

5.5 Indirect effects of acidification and nitrogen nutrient enrichment

5.6 Other effects

Page 5-7 line 29: Should this read 'Inorganic aluminum...?'

Page 5-18 lines 19 and 20: Reword as "...ecosystems causes fertilization of trees and grasslands accelerating growth in some species...."

The words "inadvertent" and "unnatural" seemed inappropriate to me.

Page 5-19 line 9: Change to "When N increases to...."

Page 5-20 lines 24 and 25: N is also retained in biomass.

Page 5-37 lines 27 and 28: I would change "many forest ecosystems" to 'sensitive forest ecosystems'. There are many forest ecosystems that not show any adverse effects from this level of N deposition for a long time (if ever).

Most of the Tables should be used in their respective Sections and associated with some descriptive text. Table 5.7-1 is appropriate to the Summary and Findings section, and I would retain it. It would be helpful, however, to add a column to Table 5.7-1 to show how the experimental levels of N&S exposure actually relate to current N or S deposition rates. In many cases the experiments use N deposition rates that are an order of magnitude above current levels.

Draft Text for an Answer to ISA Charge Question #10:

Several additional effects are discussed, including mercury methylation, direct gas-phase effects on foliage, and N₂O as a greenhouse gas. How well does the draft ISA characterize the evidence on these topics?

Designating the direct effects of gas phase NO_x and SO_x as an additional effect is inconsistent with prior AQCDs that appropriately delineated direct versus indirect effects of the defined criteria pollutants. I would argue that the ISA should return to this format and characterize gas phase effects of nitrogen and sulfur oxides as direct effects that are seldom expressed under current atmospheric conditions. That discussion could be logically followed by a discussion of the indirect effects of acidification, nitrogen nutrient additions, sulfur-induced mercury methylation, the production of the greenhouse gas N₂O, the production of tropospheric ozone, the contributions of NO_x and SO_x to visibility issues, and the interactive influence of global anthropogenic N additions on carbon sequestration within natural ecosystems. The last item on the stimulation of ecosystem carbon sequestration is not currently discussed within the ISA, but should be considered as an addition.

Some references related to N deposition and carbon sequestration:

- Grace J (2004) Understanding and managing the global carbon cycle. *Journal of Ecology* 92:189-202.
- Hyvonen R, Agren GI, Linder S, et al. (2007) The likely impact of elevated [CO₂], nitrogen deposition, increased temperature and management on carbon sequestration in temperate and boreal forest ecosystems: a literature review. *New Phytologist* 173:463-480.
- Korner C (2000) Biosphere responses to CO₂ enrichment. *Ecological Applications* 10:1590-1619.
- Makipaa R, Karjalainen T, Pussinen A, et al. (1999) Effects of climate change and nitrogen deposition on the carbon sequestration of a forest ecosystem in the boreal zone. *CANADIAN JOURNAL OF FOREST RESEARCH* 29:1490-1501.
- Nadelhoffer KJ, Emmett BA, Gundersen P, et al. (1999) Nitrogen deposition makes a minor contribution to carbon sequestration in temperate forests. *Nature* 398:145-148.
- Norby RJ (1988) Nitrogen deposition: a component of global change analyses. *New Phytologist* 139:189-200.
- Oren R, Ellsworth DS, Johnsen KH, et al. (2001) Soil fertility limits carbon sequestration by forest ecosystems in a CO₂-enriched atmosphere. *Nature* 411:469-472.
- Schindler DW, Bayley SE (1993) The biosphere as an increasing sink for atmospheric carbon - estimates from increased nitrogen deposition. *Global Biogeochemical Cycles* 7:717-733.

Dr. Dale Johnson

The quality of the review is very mixed, especially in Section 4. The first part is fraught with inaccuracies, misconceptions, and bias, the last part is even handed and complete. Indeed, there seems to be a real dichotomy between two different authors who seem to have written this section, and the second one has a much more complete and balanced view of the situation. Again, I stress the need for looking at all points of view – which in total this document does, but only in certain parts and these parts do not adequately make their way into the summary sections. The focus is on the negative effects, which leaves this document open for severe criticism once it is released. There are several peer-reviewed publications that discuss and even demonstrate the possibility that increased N deposition has or will increase terrestrial ecosystem C sequestration. Below I cite some examples:

LeBauer, D.S., and K.K. Treseder. 2008. Nitrogen limitation of net primary productivity in terrestrial ecosystems is globally distributed. *Ecology* 89: 317-379.

This is a meta analysis of fertilizer studies that investigated latitudinal trends in response to N. They note that most fertilizer applications differ from atmospheric deposition in that they are greater in quantity (at least over the short term) and pulse-like in time. They also fully recognize the potentially negative effects of too much N. The last sentence of their Conclusions reads: "Increasing N deposition, particularly in the most rapidly developing regions, is likely to further stimulate global NPP and slow the accumulation of atmospheric CO₂."

Magnini, F., Mencuccini, M., Borghetti, M., Berbigier, P., Beringer, P., Delzon, S., Grelle, A., Hari, P., Jarvis, P.G., Kolari, P., Kowalski, A.S., Lankreijer, H. Law. B.E., Lindroth, A., Loustau, D., Giovanni, M., Moncreiff, J.B., Rayment, M., Tedeschi, V., Valentini, R. and Grace, J. 2007. The human footprint in the carbon cycle of temperate and boreal forests. *Nature* 447: 848-850.

A quote from their abstract: "After the confounding effects of disturbance have been factored out, however, forest net carbon sequestration is found to be overwhelmingly driven by nitrogen deposition, largely the result of human activities."

Pregitzer, K.S., A.J. Burton, D.R. Zak, and A.F. Talhelm. 2008. Simulated chronic nitrogen deposition increases carbon storage in Northern Temperate forests. *Global Change Biology* 14: 142-153.

This is a field study where applications of N at 30 kg ha⁻¹yr⁻¹ over two decades produced greater C sequestration in surface soils and live woody tissues in northern hardwood forests in Michigan.

For the combined effects of CO₂ and N:

Hungate, B.A., J.S. Dukes, M.R. Shaw, Y. Luo, and C.B. Field. 2003. Nitrogen and climate change. *Science* 302: 1512-1513.

The authors point out that model estimates of C sequestration due to elevated CO₂ are probably greatly inflated because they do not account for N limitation. They calculate some projected increases in N deposition and estimate how much this could facilitate the modeled estimates of CO₂-enhanced C sequestration and it falls far short. So the point is that models that do not incorporate N overestimate C sequestration and also implies that more N will lead to more C sequestration.

Again, I must emphasize that this is meant to provide the other side of the N deposition issue and in no way negates the well-documented negative effects of excessive N deposition on terrestrial ecosystems. The problem with nitrogen in terrestrial ecosystems is that there is a very short plateau between deficiency, where increased growth with increased N inputs will occur, and excess, where many negative effects such as soil and water acidification and water pollution commence.

Specific comments:

p. 1-2, lines 5-22: Here I ask my recurring question: why the complete focus on adverse effects? Nitrogen is the limiting nutrient for most terrestrial ecosystems and therefore there is the distinct possibility of beneficial effects as well. Again, I do not advocate excusing air polluters on this basis, but I think a fair and complete assessment requires that this side be discussed. To fail to do so risks losing credibility for the entire effort.

p. 4-2, Title: The very title of this section clearly shows an bias toward adverse effects and such a bias is completely unacceptable in a document that purports to be scientifically objective.

p. 4-3, lines 7-8: It is very hard to imagine that increasing N deposition will fail to cause “change” in any ecosystem, whether it be beneficial or harmful change. I think there is a need to be more specific here.

p. 4-7, line 24: Acid cations that build up during soil acidification include, importantly, Al³⁺ as well as H⁺.

p. 4-7 line 25: Soil acidification IS a natural process. There is no doubt of this. Extremely acidic soils can be found in pristine parts of the world. You should elaborate on such processes here – carbonic acid, organic acids, plant cation uptake.

p. 4-7, line 28: Where have decreases in pH attributable to acidic deposition been found in the US? You should cite references here.

p. 4-8, line 6: The B horizon lies below the Oa horizon? Since when? This can happen in some cases, but is the exception rather than the rule. What happened to the A and E horizons? Does this author know anything about soils and soil genesis?

p. 4-8, lines 15-17: Actually, the situation with hardwoods is much more complicated than that and this statement is misleading. Hardwoods, by taking up larger amounts of Ca, actually acidify sub surface soil horizons more than conifers to even though they may enrich surface horizons with Ca by litterfall recycling. There is a classic paper by Alban (1982) that clearly show this and should be cited here. Also, an excellent source of information is the review paper by Stone (1975).

p. 4-8, line 28 and throughout: “SO₄⁺”? Since when is sulfate a cation? This is an embarrassing mistake. Sulfate is an anion: SO₄²⁻

p. 4-10, lines 5-6: Some oil crops have S demands that are in line with S deposition.

p. 4-10, lines 25-31: There are MANY other references besides Sullivan et al 2004 that show this.

p. 4-11, line 30: Add this to the list: “3) greater growth causing more cation uptake and therefore more soil acidification”

p. 4-12, lines 2-9: Ammonium is also acidifying: it is either taken up by plants or microbes, thereby releasing H⁺, or is nitrified, creating nitric acid.

p. 4-14, lines 17-31: This is a good description of the leaching and acidification processes.

p. 4-15, lines 1-13: There are some significant problems with this section. First of all, it is stoichiometrically impossible for the soil changes described by Bailey et al to have been caused by any known level of acidic deposition – I wrote a letter to the editor about this and felt that their reply was inadequate (Johnson, 2006). I insist that this be included in the discussion. Secondly, there are several examples in the literature – even an entire book – written about studies where plant cation uptake as well as atmospheric deposition has caused substantial soil acidification (Johnson and Todd, 1990; Richter and Markewitz, 2001; Trettin et al., 1999). This review of the soil acidification literature for the US is totally inadequate and misleading.

p. 4-16, lines 17-26: In nature, the rate of soil weathering seldom exceeds the rate of acidification – that is why soil naturally acidify. So it is not realistic to assume that soil weathering will aid in the recovery of acidified soils.

p. 4-18, Figure 4.2-2: It appears that you have the wrong figure here. I see a soil profile, not an in situ bioassay.

p. 4-23, line 16: “lowered or INCREASED plant productivity”

p. 4-24, lines 2-3: why spell out potassium and sodium but not Ca and Mg?

p. 4-24, line 30: Al is not toxic to all tree roots. Some plants tolerate it and like acidic soils.

p. 4-26, line 12: Do you mean Nitrogen to Carbon ratio? If so, the usual expression is carbon to nitrogen ratio.

p. 4-31, lines 10-13: I thought that the red spruce decline was largely attributed to climatic factors. What about Art Johnson's work?

p. 4-37, lines 5-10: How about some of the N-loving invasive grasses in the southwestern US like Bromus species? Edie Allen has shown that N deposition clearly facilitates this in southern California.

p. 4-43, line 22: Do you mean cation here?

p. 4-43, lines 25-26: In the study of Lawrence et al, did H^+ and Al^{3+} decrease to make up the difference between base cation decreases and mineral acid anion decreases? Something surely had to, otherwise charge balance was not maintained.

p. 4-44, lines 4-5: How were strongly acidic organic anions estimated?

p. 4-63: A discussion of capacity/intensity and anion mobility concepts for water acidification is needed here. Namely, that a strong acid anion such as sulfate or nitrate passing through an acid soil can mobilized H^+ and Al^{3+} with no delay because these acid cations are most available on exchange sites to balance the anions. This reaction does not require soil change and is instantly reversible if strong acid anion inputs cease. This is quite a different matter from the case where soils acidify. An already acidic soil, whether by natural acid production or by acid rain, is a necessary but not sufficient condition for the acidification of soil water – strong acid anions are also needed.

p. 4-84, lines 13-23: This is very interesting – we did not expect that soils of the northeast would have such buffering with respect to sulfate. Very important point.

p. 4-91, Figure 4.2-17: The y-axis legends on this figure are messed up.

p. 4-104, line 30: “unnatural growth rates?” Again, this reflects the negative bias that seriously detracts from this entire document. Just say increased growth rates and let the reader decide if it is good or bad.

p. 4-105, lines 16-17: Once again, this statement reflects the negative bias that seriously detracts from this entire document. I suspect that you would also be looking for negative effects if N limitations were increased rather than decreased in the endless search for negative consequences. Please let us simply state what we think will happen and leave out these value judgements.

p. 4-106, lines 9-10: At LAST! We see the mention of potential beneficial effects.

p. 4-111, lines 3-17: Once again, there is too much negative bias here. I raise once again the issue of global C and the potential for N pollution to make the terrestrial C balance better (Kauppi et al., 1992; Magnini et al., 2007).

p. 4-121, lines 12-32: This is as close to a balanced presentation as I have yet seen in this document. It needs to be up front along with all the potential negative effects.

p. 4-122, lines 15-16: Of course adding the limiting nutrient causes the next most limiting nutrient to become limiting – everyone who has ever worked with fertilization knows this. Why is this a bad thing? Is it healthier for a forest to be N limited than Ca, K, or Mg limited? Once again, an unnecessary value judgement here.

p. 4-123-124: At last we see the new article by Magnini and a balanced discussion about it. I think this kind of alternative view needs to be more up front as well.

p. 4-126: Good discussion of potential effects on grasslands and invasive species problems in relation to N.

p. 4-127-128: Again, a very good discussion of potential N effects on arid and semi-arid lands. N inputs in these areas could be a real problem and there is not much to be seen in the way of benefits.

p. 4-161-162: Good discussion of the non-acidifying effects and biological role of sulfur.

p. 5-6, lines 19-31: Some of that good discussion and balanced treatment seen on the last pages of section 4 need to make their way into this summary.

p. 5-7, line 29: Aluminum is toxic to SOME tree roots.

p. 5-8: Again, Some of that good discussion and balanced treatment seen on the last pages of section 4 need to make their way into this summary.

p. 5-12, lines 2-4: This is a very important yet seldom recognized point about surface water vs soil acidification.

p. 5-18, lines 19-20: Once again, why must we say unnatural? Why not just INCREASED growth rates? And then go on to elaborate about how you do not want this to happen in certain places (if indeed you do want to maintain N deficiencies).

p. 5-21: Good summary about N effects on growth, both positive and negative.

References:

Bailey, S. W., S. B. Horsley, and R. P. Long. 2005. Thirty years of change in forest soils of the Allegheny Plateau, Pennsylvania. *Soil Sci. Soc. Am. J.* 69:681-690.

- Johnson, D.W. 2006. Comments on “Thirty years of change in forest soils of the Allegheny Plateau, Pennsylvania.” *Soil Sci. Soc. Amer. J.* 69: 2077.
- Johnson, D.W., and D.E. Todd. 1990. Nutrient cycling in forests of Walker Branch Watershed: Roles of uptake and leaching in causing soil change. *J. Environ. Qual.* 19: 97-104.
- Kauppi, P.E., Mielikäinen, K. and Kuusela, K., 1992. Biomass and carbon budget of European Forests, 1971 to 1990. *Science*, 256: 70-74.
- Magnini, F., Mencuccini, M., Borghetti, M., Berbigier, P, Beringer, P., Delzon, S., Grelle, A., Hari, P., Jarvis, P.G., Kolari, P., Kowalski, A.S., Lankreijer, H. Law. B.E., Lindroth, A., Loustau, D., Giovanni, M., Moncreiff, J.B., Rayment, M., Tedeschi, V., Valentini, R. and Grace, J. 2007. The human footprint in the carbon cycle of temperate and boreal forests. *Nature* 447: 848-850.
- Richter, D.D., and D. Markewitz. 2001. Understanding soil change. Soil sustainability over millennia, centuries, and decades. Cambridge University Press.
- Stone, E.L. 1975. Effects of species on nutrient cycles and soil change. *Philos. Trans. R. Soc. Lond. Ser. B Biol. Sci.*, 721: 149-162.
- Trettin, C.A., D.W. Johnson, and D.E. Todd, Jr. 1999. Forest nutrient and carbon pools: a 21-year assessment. *Soil Sci. Soc. Amer. J.* 63: 1436-1448.

Dr. Donna Kenski

Overall, this was a fine job summarizing a lot of data. The quality varied from section to section, but most of the relevant information was here. Nevertheless there were some significant shortcomings that should be addressed in future drafts. Most of my comments address the presentation of air quality and emissions data, per Charge Questions 1-3.

The chemistry discussion is adequate. Most of the air quality issues were covered in Chapter 3, and I found them lacking in several ways, especially with respect to the nitrogen species. Section 3.1 needs a map of NO_x emissions density (all sources) and a comparison, at least nationally but preferably regionally, of the relative importance of each of the anthropogenic and biogenic sources to total NO_x emissions. We're told that soil contributes 10% of NO_x emissions globally, and 26% of NO in Illinois, but no estimates are given for soil NO_x contribution to US emissions either nationally or regionally. Similarly for biomass burning. The statement that increased wildfires will make emissions from this sector increasingly important should be substantiated with quantitative data. This information is too important to relegate to the Annex, and Table 3.11-2 puts sources in priority order but is not quantitative. Table AX2-1 contains the relevant information, and a shortened version of it, edited to eliminate the many small sources and include only major categories would add significantly to this section. The section also needs a description of NO_x emission trends, parallel to the discussion of SO_x emission trends in Sec. 3.2. While changes in NO_x have not been as dramatic as those for SO_x, they are documented and the data are readily available. In addition, some information about future-year emissions projections would be nice, along with a brief summary of recent and expected controls on NO_x and SO_x sources. These analyses have been done by EPA already. Another significant hole in the emissions discussion was NH₃. Although we're told that the NEI underestimated NH₃ emissions by a factor of 2 or 3, we're never given an estimate of what those emissions are (underestimate or not).

Section 3.6.1, and especially p. 3-15, lines 9-15 is somewhat misleading in its description of the monitoring networks. Sure we have lots of ozone monitors, but ozone monitors don't tell us much about NO_x or SO_x or NH₃. I don't understand why the authors of this section decided to show us plots of ozone and PM_{2.5} air quality but not plots of SO₂, NO_x or NH₃ concentrations. While it is true as stated in lines 11-13 that NO_x and SO_x are often monitored at the same sites, the number of NO_x and SO_x monitors is far less than for ozone or PM_{2.5}. More relevant would be a plot of just NO₂ and SO₂ monitors, which would make immediately apparent the significant gaps in spatial coverage of measurements for these gases. Figures 3.6-1 and 3.6-2 leave the reader with the impression that the network of NO_x/SO_x monitors is much more dense than it really is. Including NATTS, HAPS, CO, and lead sites in Fig. 3.6-2 only obscures the matter further. It may also be worth noting that monitors for both NO_x and SO_x have been cut from the networks in recent years because their main purpose is perceived to be for comparing to the primary NAAQS. As ambient concentrations for both NO_x and SO_x have fallen well below the standard throughout the US, the monitors are seen as a lower priority and unnecessary expense. At the same time, monitoring budgets have been cut, leading to monitor shut downs.

Another shortcoming of this section is that it does not address urban-rural differences. Figure 3.6-3, for example, is of limited usefulness in assessing the composition of aerosol that impacts the sensitive, mostly rural regions identified later; differences in urban and rural aerosols have been noted by many. A similar plot, but based on IMPROVE data, would be an improvement, or a plot that shows both urban and rural composition. The majority of monitors, both gas and particle, are in urban areas; rural NO_x and SO_x monitors are rare. How does the lack of rural NO_x and SO_x monitors affect estimates of N deposition?

Figures 3.7-2 and 3.7-3, which compares ambient SO₂ and SO₄ concentrations in the early 1990s with more recent data, are very effective, but the text or caption should note whether Fig. 3.7-3 is showing SO₄ concentrations in particulates or in rainfall – its not clear which. A similar set of plots should be shown for NO_x and NH₃.

I didn't like Fig. 3.7-4 when it showed up in the SO_x primary ISA (as I recall from discussions, none of the panel did) and I still don't like it. It is not effectively communicating information about SO₂ concentrations; replotting on a log scale would make it much more informative. Especially in this context, the extreme values are less informative than the rest of the distribution.

Figures 3.8-1 thru 3.8-4 are also ineffective. It is difficult to distinguish meaningful differences by comparing the size of the pies in the upper and lower plots, and also to tell if there have been changes in the geographic distribution of the various forms of S and N deposition. NADP produces a much more effective set of plots, at least for wet deposition, similar to Fig. 3.7-2; perhaps those could be substituted here (see <http://nadp.sws.uiuc.edu/isopleths/annualmaps.asp>) or these could be reworked to show the data in a similar fashion. At least change the pies to bar charts as in Fig. 3.2-1.

Section 3.6.4 on satellite observations was too brief to be useful. It seems clear that the technology holds great promise; it would be helpful to see some support of that notion, rather than just have a laundry list of satellites without any indication of their strengths and weaknesses. Is there useful data for our purposes (N and S deposition) being produced by the satellites now? If so report it here.

Section 3.6.5 could be removed to Annex 2.

This chapter should have left us with an understanding of the relative importance of NO_x, NH₃, and organic nitrate deposition nationally and regionally, but at the end I still felt unable to assess those except in a qualitative way.

Ultimately, the question we want to answer is, what kind of secondary standard would be adequately protective of our sensitive ecosystems. Here I think the ISA was not very helpful. I was searching for information that explicitly linked ambient concentrations or emissions with deposition and more importantly with acidification effects. How are ambient concentrations related to deposition? Are these essentially equivalent? Can we compare (using GIS tools or other quantitative techniques) the distribution of concentrations (or deposition) to the distribution of sensitive ecosystems?

Some other general comments:

The various chapters and subsections make occasional mention of shortcomings in the data and areas that are poorly understood, but these shortcomings aren't collectively addressed anywhere. These data gaps and research needs should be explicitly discussed and summarized in the conclusions. Doing so would help us better understand some of the limits to our knowledge and also hopefully help EPA target research efforts and support future reviews.

Another concern I have is that NH_x isn't addressed comprehensively here because the focus is NO_x and SO_x , and it may not be comprehensively discussed in the PM ISA, because the focus there is PM. When or where does NH_x get its due? If we always consider it as a smaller part of something else, it may never get dealt with adequately. I appreciate that NH_x 's contributions to deposition and acidification were discussed here at some length, but we seem in danger of marginalizing its very important role in nitrogen and sulfur deposition because it is a lesser player, without doing the research to adequately characterize its role.

More specific comments:

p. 2-29 line 6-7 The concluding sentence, that coverage of monitoring networks is thin over large expanses of the US, is true, but not a conclusion that follows from the previous discussion, which is only about the adequacy of the monitoring techniques. It is a statement more appropriate for Sec. 3.6.1.

p.3-7 line 7-9 overrepresentation is a poor word choice. Perhaps higher density would be better.

p. 3-13, eqn 3.5-4 Isn't the second = sign supposed to be a + ?

p. 3-16, caption to Fig. 3.6-2 should be ..majority *of* routine...

p. 3-35, caption to Fig. 3.9-1 should read ...25th and 75th quartiles...

p. 4-7 Throughout this Section (4.2) the concept of base saturation is discussed and a few benchmark figures (base saturation less than 20%, for example) are mentioned, but it does not explain how base saturation is quantified until p. 4-24. A brief definition at the beginning of this section would be a useful addition since it may not be familiar to the many in the atmospheric community.

pp. 4-7 - 4-191 Somehow in this entire chapter, SO_4^{2-} morphed into SO_4^+ . Search and replace gone awry? Very disconcerting.

pp. 4-9, 4-18 Figures 4.2-1 and 4.2-2 have been switched. The captions are correct but the figures themselves are reversed.

p. 4-24 Later in the document there are maps of sensitive areas (although not enough—much more of the data could/should have been summarized graphically). Any available to show soil base saturation, or Ca:Al ratios, which might be helpful here?

p. 4-24 Change in terms soil water -> soil solution is confusing. Are these the synonyms or is there some subtle difference?

p. 4-26 line 12 Calcium should be Carbon

p. 4-64, section 4.2.4.2 This section suffers from a lack of integration. It's difficult to assess the extent and distribution of these sensitive areas without some graphical help. Figure 4.2-12 is a start, but only tells part of the story; Figure AX4.3-2 does a much better job showing the location and extent of acidification. Also, acidified streams and lakes are sometimes mentioned separately and sometimes together; it is not clear whether regions with acidified streams necessarily also have acidified lakes, and vice versa.

p. 5-16, lines 5-14 Although this section is supposed to be about recent trends in acidification recovery, this last bullet doesn't say anything about recovery and seems out of place. The parent section 4.2.4.2.1 emphasizes that this WSA survey is summer data and therefore biased low, but that caveat is not repeated here, as it should be. These results also don't jive with later data (i.e., on p. 5-17, 66% of Adirondack streams currently have high Al). It would be useful to have a graphical assessment of the geographic extent of acidification either here or as suggested earlier, in section 4.2.4.2. Figure AX4.3-2 is very helpful in this regard.

p. 5-36 lines 19-23 Absolutely true that regulatory networks cannot adequately characterize regional heterogeneity (on a scale that matches that of the sensitive ecosystems) and hotspots. But the statement about hotspots identified at research sites was not discussed in the text, except in reference to Hg. If this is documented it should be discussed in the body of the ISA, not mentioned first in the summary.

p. 5-36, line 25 lead should be led

p. 5-38 This section comes to an abrupt end; is something missing?

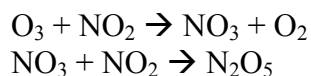
Dr. Naresh Kumar

Information provided in the Integrated Science Assessment document on atmospheric chemistry and physics, air quality, and deposition and exposure is concise, but sufficiently comprehensive. Specific comments are:

Page 2-2, Lines 6 & 7: Nitric acid doesn't contribute to the acidity of particles, so there is a need to reword the sentence.

Page 2-2, Lines 23 & 24: It would be meaningful to mention the contribution to NO_x emissions from mobile sources and electric utilities (e.g., According to NEI 2003, 55% of NO_x emissions were from mobile sources and 22% were from electric utilities).

Page 2-5, Line 7: The following reactions should be added before Equation 2.2-2



Page 2-5, Line 15: The nitrate radical, NO₃ should be included as part of the photolysis.

Page 2-6, Lines 7 thru' 16: The paragraph should also mention the negative radiative forcing caused by nitrate particles.

Page 2-6, Line 28: Add "from the free troposphere" at the end of the sentence.

Page 2-7, Line 9: Add "except at lower temperatures" at the end of the sentence.

Page 2-8, Line 13: There is no SO₄²⁻ in Equation 2.2-15.

Page 2-8, Line 22: "diatonic" should be "diatomic".

Page 2-9, Line 31: Change "high" to "moderate"

Page 2-9, Line 32: After "in the aqueous phase in cloud droplets", add "by reaction with H₂O₂, O₃ or O₂ through catalysis by Fe and Mn.

Page 2-10, Line 7: After "highly soluble", add "as well as hygroscopic".

Page 2-11, Line 3: "Methane sulfuric acid" should be "methanesulfonic acid".

Page 2-12, Figure 2.3-1: Include Fe, Mn and O₂ where reaction from S(IV) to S(VI) is shown.

Page 2-13, Line 18: Add manganese (Mn) to the list of metals that can catalyze oxidation by O₂.

Page 2-17, Line 17: Include Edgerton et al., 2006 and Edgerton et al., 2007 in the list of references on newer methods for measuring nitrate, ammonia and ammonium. SEARCH network has been in place since 1999 and has been widely used by the community, but there is no mention of this network anywhere in the ISA document.

Page 2-21, Line 12: H₂SO₄ should be changed to SO₂.

Page 2-24, Line 22: Add “for IMPROVE and thermal-optical transmittance (TOT) for STN” at the end of the sentence.

Page 2-25, Line 2: Change “well correlated” to “moderately well correlated”.

Page 2-26, Line 1: Remove NH₄NO₃ from the list.

Page 2-26, Line 6: After “PM₁₀”, add “, partly because NO₃⁻ contributes smaller fraction to PM₁₀ and partly because NO₃⁻ is present in a non-volatile form, such as NaNO₃, in the coarse mode.”.

Page 3-1, Lines 23 & 24: Electrical utilities and various industries account for about one-third of anthropogenic NO_x emissions (not roughly half).

Page 3-20, Line 13: Add description of the SEARCH network. Here is the suggested description to choose from:

Southern Company and EPRI funded SEARCH program has provided a highly instrumented eight-station network since 1998 (and continuing till at least 2010) in the states of AL, FL, GA and MS (Hansen et al., 2003). At present, the suite of measurements made at all sites includes:

- 1. 24-hr PM_{2.5} filter samples, analyzed for mass, ions (sulfate, nitrate, ammonium), organic carbon(OC), elemental (black) carbon (EC or BC), and elements as measured by X-ray fluorescence (XRF);*
- 2. 24-hr PM_{Coarse} mass, ions, and XRF elements;*
- 3. 24-hr gaseous ammonia as collected with an annular denuder;*
- 4. continuous (minute to hourly) PM_{2.5} mass, OC, EC, ammonium, nitrate, and sulfate; light scattering and light absorption;*
- 5. continuous gaseous ozone, nitric oxide, nitrogen dioxide, total oxidized nitrogen (NO_y), nitric acid, carbon monoxide, and sulfur dioxide; and*
- 6. continuous 10-m meteorological parameters: wind speed, wind direction, temperature, relative humidity, solar radiation, barometric pressure and precipitation.*

Page 3-23, Line 22: SEARCH sites have also measured nitric acid (Zhang et al., 2006; Blanchard and Hidy, 2003) since 1998 and the measured hourly concentrations range from less than 1 ppb to more than 10 ppb.

Page 3-42, Line 14: Add “international transport” as one of the issues addressed by global CTMs.

Page 3-43, Line 17: I suggest adding Arnold and Dennis (2006) reference, as they went beyond just testing the operational model performance.

Page 3-47, Line 27: I suggest adding appropriate references to the SEARCH sites here.

References:

- Arnold, J.R. and Dennis, Robin L. (2006). Testing CMAQ chemistry sensitivities in base case and emissions control runs at SEARCH and SOS99 surface sites in the southeastern US. *Atmos. Environ.* **40** (26) 5027-5040.
- Blanchard, C.L. and Hidy, GM. (2003). Effects of changes in sulfate, ammonia, and nitric acid on particulate nitrate concentrations in the southeastern United States. *J. Air Waste Manage. Assoc.* **53** 283-290.
- 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 Manage. Assoc.* **56**, 1325-1341.
- Edgerton, Saylor, Hartsell, Jansen, Hansen (2007) Ammonia and Ammonium Measurements from the Southeastern U. S., *Atmos. Environ.*, **41**, 3339-3351.
- Hansen, D.A., Edgerton, E.S., Hartsell, B.E., Jansen, J.J., Kandasamy, N., Hidy, G.M., and Blanchard, C.L. (2003). The Southeastern Aerosol Research and Characterization Study: Part 1 – Overview. *J. Air Waste Manage. Assoc.* **53** 1460-1471.
- Zhang, Y., Liu, P., Queen, A., Misenis, C., Pun, B., Seigneur, C., and Wu, S.-Y. (2006). A comprehensive performance evaluation of MM5-CMAQ for the Summer 1999 Southern Oxidants Study episode—Part II: Gas and aerosol predictions. *Atmos. Environ.* **40** (26) 4839-4855.

Dr. Myron Mitchell

General Comments

The December 2007 version of the “Integrated Science Assessment for Oxides of Nitrogen and Sulfur – Environmental Criteria” has done a very good job in summarizing a substantial amount of information. Included within this analysis is discussion on the relevance of this information to policy decisions. There is some reference and discussion to results from European analyses. It would be interesting to make some further explicit comparisons of the success or lack of success of the European analyses including the relevance of these analyses to both interpretations and policy implications in the United States.

In the various sections, however, there is sometimes substantial repetition and overlap. Some of this is likely inevitable, but having additional cross referencing among chapters would also be helpful. Removing some of this overlap will make the document more readable and reduce the overall length. In Chapter 3 (ECOLOGICAL EXPOSURES TO OXIDES OF NITROGEN AND SULFUR, AND TO AMMONIA AND AMMONIUM) some further discussion of the issues related to model parameterization, assumptions and differences in the predictions of concentrations and deposition would be helpful in placing results in the context of the overall confidence levels of predicted values both across temporal and spatial scales.

In Chapter 4 (EFFECTS OF ACIDIFICATION AND NITROGEN ENRICHMENT ON ECOSYSTEMS AND OTHER WELFARE EFFECTS) more attention should be given to the potential importance in at least some watersheds of other sources of sulfate that will affect the recovery from acidification. These sources can include organic sulfur mineralization and the weathering of sulfur bearing minerals.

In Chapter 5 (FINDINGS AND CONCLUSIONS), this information should be converted to an Executive Summary in which the salient features of the document are articulated.

Inclusion in the assessment of the impact of total reactive nitrogen would help strengthen the importance of all reactive nitrogen chemical species including ammonium to the total nitrogen loading of ecosystem.

More Detailed Comments

Page	Comment
xx	Change to: Dr. Myron Mitchell, Distinguished Professor and Director of Council on Hydrologic Systems Science, College of Environmental and Forestry, State University of New York, Syracuse, NY
xxxv	Change to: The removal of gases and particles from the atmosphere to surfaces by rain or other forms of precipitation

- 2-2 Change to: contributing to the acidity of cloud, fog, and rain water and ambient particles in the atmosphere.
- 2-3 Change to: few hours during summer days of high photon flux to roughly 24 h during winter with less
- 2-4 Change to:
2002). The mechanisms for transporting the NO_x precursors, the factors controlling the
- 2-6 Change to: on the conventional 100-year time horizon of ~296 – i.e., N₂O is nearly 300 times more effective on a per molecule basis for trapping heat in the atmosphere than carbon dioxide (CO₂)(IPCC,
- 2-13 Indicate location of “Mace Head” in line 3.
- 2-18 Be more explicit in what is the interference by NO_z compounds in line 1.
- 2-20 Reword to clarify the issue related to: also lead to artifact volatilization and associated positive bias in [HNO₃] measured downstream
- 2-20 Would this be an appropriate place to discuss issues related to differences between CASTNET and the Canadian (CAPMoN) dry deposition comparisons. I believe most of the differences are associated with the dry deposition modeling versus the actual measurements of gaseous chemical species.
- 2-22 Figure 2.6-1 needs to provided with better resolution of the contour lines.
- 2-24 Should Teflon be indicated to be a registered name? ®
- 3-1 Delete: (The category label for NH₃ and NH₄⁺ is NH_x.)
- 3-1 Substitute “approximately” for “roughly” here and elsewhere in the document.
- 3-2 Change to: Where N is in excess of biotic demand, gaseous N emissions increase by microbial transformation.
- 3-2 The issue related to “Although N₂ is not reactive in the troposphere, N₂O is a greenhouse gas (GHG) with a significant global warming potential (GWP) from its direct radiative forcing and from its...” maybe should be linked to the discussion in Chapter 2.
- 3-3 Change to: Emission rates of NO from cultivated soils depend chiefly on N fertilization levels
- 3-3 Change to: can have wide variation around this value

- 3-3 Change to: N in plants is present mostly as amine (NH₂) groups in amino acids.
- 3-3 Change to: will make emissions from this source increasingly important
- 3-8 This statement needs clarification: Emissions of SO₂ from burning vegetation are generally in the range of 1 to 2% of the biomass burned (e.g., Levine et al., 1999).

This would suggest that 1 to 2% of biomass gets converted to SO₂. This seems to high with respect to the average S concentration in vegetation. Does this study suggest that 1 to 2% of the S in biomass is converted to SO₂?
- 3-9 Be more explicit about which pattern in the statement: Anthropogenic emissions of NH₃ show a strikingly different pattern from those of NO_x or SO₂.
- 3-10 Does the statement “Results from some more recent emissions evaluation studies have been mixed, with some studies showing agreement to within ±50%” imply that some of these studies show that agreement is greater than 50%?
- 3-21 Figure 3.6-7. Routinely operating North American precipitation and surface water networks: Upper left, Canadian Air and Precipitation Monitoring Network (CAPMoN); upper right, Integrated Atmospheric Monitoring Deposition Network (IADN); bottom, National Atmospheric Deposition Monitoring Program (NADP) with TIME/LTM surface chemistry sites.

This figure shows a symbol of N-saturated forests. What is the source of this designation? What is the coverage associated with the symbol? Other Ecological Resources are also noted, but the source of these designations is not included in the figure legend.
- 3-22
- 3-23 Some further discussion of differences of these European monitoring efforts compared to those in the U.S. may be helpful. Also, some discussion on differences with the Canadian monitoring efforts should be included.
- 3-47 Change to: parameterizations of atmospheric chemical and physical processes in models.
- 3-48 Change to: In a watershed, everything that is deposited in its area
- 4-1 Change: Structure refers to the species richness, abundance, and community composition that ultimately relate to ecosystem biodiversity.

To something like: Structure may refer to a variety of measurements including the species richness, abundance, community composition as well as landscape attributes. The biotic components compose the biodiversity of an ecosystem.

- 4-1 Change to: Competition among and within species and tolerance to environmental stresses are key elements of survivorship
- 4-1 Change to: Function refers to the suite of processes and interactions among the ecosystem components and their environment that such as nutrient and energy flow as well as other attributes including water dynamics and the flux of trace gases.
- 4-2 Change to: related to functions of energy flow and nutrient cycling
- 4-2 Change to: Energy moves from one organism to another through food webs, until it is ultimately released as heat. Nutrients and water can be recycled. Air pollution alters the function of ecosystems when elemental cycles or the energy flow is altered. This alteration can also be manifested in changes in the biotic composition of the ecosystem.
- 4-6 The use of “we” seems awkward in this type of discussion. I would suggest rewording.
- 4-8 It may also be useful to include the term “forest floor” when discussing the O horizon of forest soils.
- 4-8 For the documentation of soil acidification the following reference should be included: Sullivan, T.J., I.J. Fernandez, A.T. Herlihy, C.T. Driscoll, T.C. McDonnell, N.A. Nowicki, K.U. Snyder, and J.W. Sutherland. 2006. Acid-base characteristics of soils in the Adirondack Mountains, New York. Soil Science Society of America Journal 70: 141-152.
- 4-11 Not only is the fate of carbon-bonded sulfur an issue, but also the fate of ester sulfate needs to be considered.
- 4-11 This might be the place to at least mention that sulfate isotopic have indicated the potential for the net mineralization of organic S contributing to sulfur imbalances for watersheds. This has been found in studies both the United States and Europe. Some relevant references would include:
- Novák, M. M. J. Mitchell, I. Jacková, F. Buzek, J. Schweigstillová, L. Erbanová, R. Prikryl and D. Fottová. 2007. Processes affecting oxygen isotope ratios of atmospheric and ecosystem sulfate in two contrasting forest catchments in Central Europe. ES&T 41(3): 703-709. DOI: 10.1021/es0610028
- Likens, G.E., C.T. Driscoll, D.C. Buso, M.J. Mitchell, G.M. Lovett, S.W. Bailey, T.G. Siccama, W.A. Reiners, C. Alewell. 2002. The biogeochemistry of sulfur at Hubbard Brook. Biogeochemistry 60:235-316.

Gbondo-Tugbawa, S.S., C.T. Driscoll, M.J. Mitchell, J.D. Aber and G.E. Likens. 2002. A model to simulate the response of a northern hardwood forest ecosystem to changes in S deposition. *Ecological Applications* 12:8-23.

In addition, some mentioned should be included about the potential role of weathering reactions for contributing to sulfate in some watersheds. A good example is:

Shanley, J.B., B. Mayer, M.J. Mitchell, R.L. Michel, S. Bailey and C. Kendall. 2005. Tracing sources of streamwater sulfate during snowmelt using S and O isotope ratios of sulfate and ³⁵S activity. *Biogeochemistry* 76:161–185

4-14 The is also strong evidence of the importance of winter time processes and the possible linkage to climate in affecting nitrate losses. See for example:

Eimers, M.C., Buttle, J.M., Watmough, S.A., 2007. The contribution of rain-on-snow events to annual NO₃-N export at a forested catchment in central Ontario, Canada. *Applied Geochemistry*, 22: 1105-1110.

Campbell, J.L., M. J. Mitchell, P. M. Groffman, L. M. Christenson. 2005. Winter in northeastern North America: An often overlooked but critical period for ecological processes. *Frontiers in Ecology* 3(6):314-322.

Park, J, M.J. Mitchell, P.J. McHale, S.F. Christopher and T.P. Myers. 2003. Interactive effects of changing climate and atmospheric deposition on N and S biogeochemistry in a forested watershed of the Adirondack Mountains, New York State. *Global Change Biology* 9:1602–1619.

4-20 to
4-22 There is evidence that episodic acidification associated with droughts and the mobilization of sulfate is also important. This has been shown for studies in Canada and the northeast U.S. See for example:

Eimers, M.C., Watmough, S.A., Buttle, J.M., Dillon, P.J., 2007. Drought-induced sulphate release from a wetland in south-central Ontario. *Environmental Monitoring and Assessment*, 127: 399-407.

Eimers M.C. and Dillon P.J. 2002. Climate effects on sulphate flux from forested catchments in south-central Ontario. *Biogeochem.* 61: 337–355.

Mitchell, M.J. K.B. Piatek, S. Christopher, B. Mayer, C. Kendall and P. McHale. 2006. Solute sources in stream water during consecutive fall storms in a northern hardwood forest watershed: a combined hydrological, chemical and isotopic approach. *Biogeochemistry* 78: 217-246.

- 4-26 Should this heading be Changed to: 4.2.2.1.3 Soil Nitrogen: Carbon to Nitrogen Ratio
- Not sure if the emphasis is on Ca and if the ratio indication is appropriate.
- 4-33
& Elsewhere Change SO_4^+ to: SO_4^{2-}
- 4-39 Change to: The increasing trend in Virginia streams is presumably the result of decreased soil solution sulfate concentration and net desorption in the soil in response to decreased S deposition.
- 4-41 As indicated above (see comments for 4-14) that there is other evidence that changes in winter time conditions can affect nitrate export.
- 4-42 Other studies in the Adirondacks have also identified the importance of within lake processes in affecting nitrate losses from lake/watersheds. See for example:
- Ito, M., M. J. Mitchell, C.T. Driscoll, R. M. Newton, C.E. Johnson, and K.M. Roy. 2007. Controls on surface water chemistry in two lake-watersheds in the Adirondack region of New York: differences in nitrogen solute sources and sinks. *Hydrological Processes* 21:1249-1264.
- Ito, M., M.J. Mitchell, C.T. Driscoll and K.M. Roy. 2005. Nitrogen input-output budgets for lake-watersheds in the Adirondack region of New York. *Biogeochemistry*. 72:283-314.
- 4-44 Change to: with the exception of streams in Shenandoah National Park, Virginia, which appear to exhibit decreases in adsorbed sulfate in soils.
- 4-62 to
4-63 The discussion in this section needs to differentiate between bedrock geology and surficial geology. Surficial geology is a much more important predictor of the sensitivity to acidification than bedrock geology. Differences in surficial geology can be especially important for predicting sensitivity to acidification within a region.
- 4-73 Some of this discussion on critical loads is repetitious from previous sections although this previous section is referenced.
- 4-82 I don't believe the following statement is true for the Adirondacks: However, recent research suggests that N has accumulated in soils over time in the Adirondacks and that some forests have exhibited declining retention of N inputs. The result has been increased leaching of NO_3 to surface waters.

See for example:

Mitchell, M.J., C.T. Driscoll, S. Inamdar, G. McGee, M. Mbila, and D. Raynal. 2003. Nitrogen biogeochemistry in the Adirondack mountains of New York: hardwood ecosystems and associated surface waters. *Environmental Pollution* 123:355-364.

Mitchell, M.J., C.T. Driscoll, J. Owen, D. Schaefer, R. Michener, and D.J. Raynal. 2001. Nitrogen biogeochemistry of three hardwood forest ecosystems in the Adirondack Mountains. *Biogeochemistry* 56: 93-133.

4-112 Wetlands after droughts can also serve as nitrate sources when rewetted. See for example:

Watmough S.A., Eimers M.C., Aherne J. and Dillon P.J. 2004. Climate effects on nitrate export from forested catchments in south-central Ontario. *Environ. Sci. Technol.* 38(33): 2383–2388.

Also, some mention of the role of N-fixation should be included for wetlands; see: Hurd, T. M., D. J. Raynal, and C. Schwintzer. 2001. Symbiotic N-fixation of *Alnus incana* spp. *rugosa* in shrub wetlands of the Adirondack Mountains, New York. *Oecologia* 126: 94-103.

4-129 The following paper could be referenced showing the effect on N additions on understory communities in the Adirondacks:

Hurd, T.M., A.R. Brach, and D.J. Raynal. 1998. Responses of understory vegetation of Adirondack forests to nitrogen additions. *Can J. For. Res.* 28: 799-807.

4-162 There is fairly good evidence that for forest ecosystems with closed canopies that throughfall sulfate is a good estimate of total sulfur deposition. See: Lovett G.M., Thompson A.W., Anderson J.B. and Bowser J.J. 1999. Elevational patterns of sulfur deposition at a site in the Catskill Mountains, New York. *Atmospheric Environment* 33: 617–624.

4-162
to

4-170

The discussion on the formation of MeHg needs to emphasize the interrelationship with sulfate concentrations and the role of sulfate reducing bacterial. Such focus will help emphasize that theme of this report relates nitrogen and sulfur. The extensive discussion on Hg biogeochemistry and environmental effects can be condensed.

4-179 An evaluation of nitrate sources to waters with respect to atmospheric deposition needs to separate how nitrogen deposition loading leading to more nitrate loss

versus atmospherically deposited N being found directly in drinking water. Forest watershed level work using stable isotopes shows predominantly that almost all of the nitrate in surface and ground waters has been biotically derived. See for example:

Burns, D. A. and Kendall, C.: 2002, Analysis of $\delta^{15}\text{N}$ and $\delta^{18}\text{O}$ to differentiate NO_3 sources in runoff at two watersheds in the Catskill Mountains of New York', *Water Res. Res.* 38(9), 1–11.

Piatek, K.B., M.J. Mitchell, S.R. Silva and C. Kendall. 2005. Sources of nitrate in Adirondack surface water during dissimilar snowmelt events. *Water, Air and Soil Pollution* 165:13-35.

Kendall, C.: 1998, 'Tracing nitrogen sources and cycling in catchments', in C. Kendall and J. J. McDonnell (eds), *Isotope Tracers in catchment hydrology*, Elsevier Science, BV, pp. 519–576.

5-36
to
5-37

The conclusion section should then be shorted and include information on the overall approach of the study including important resources to be used and potential limitations.

5-3

Not sure we have good estimates for some pollutants such as ammonia. This statement seems contradictory:

Despite the aforementioned limitations, for the purposes of identifying and quantifying the atmospheric concentrations and deposition totals causing ecological effects, these measurement techniques and sampling frequencies are fully adequate. Nevertheless, the coverage of the networks is very thin over large expanses of the interior United States.

5-6

As stated previously the importance of superficial geology needs to be emphasized.

5-36
to
5-42
to
5-44

TABLE 5.7-1. SUMMARY OF NITROGEN DEPOSITION LEVELS AND THE CORRESPONDING EFFECTS OF ACIDIFICATION AND NUTRIENT ENRICHMENT ON ECOSYSTEMS.

To this table could be added the results from the Adirondack Manipulation and Modeling Program (AMMP): Mitchell, M.J., C.T. Driscoll, J. Owen, D. Schaefer, R. Michener, and D.J. Raynal. 2001. Nitrogen biogeochemistry of three hardwood

forest ecosystems in the Adirondack Mountains. *Biogeochemistry* 56: 93-133.)

This study included experimental additions of ammonium sulfate and nitric acid (14 and 28 kg N ha⁻¹ yr⁻¹). The biogeochemistry of nitrogen (N) was evaluated for three forest ecosystems [Woods Lake (WL), Pancake-Hall Creek (PHC) and Huntington Forest (HF)] in the Adirondack region. Bulk N deposition was higher at sites in the west than those in the central and eastern Adirondacks. These higher atmospheric N inputs were reflected in higher bulk throughfall fluxes of N (WL and PHC, 10.1 and 12.0 kg N ha⁻¹ yr⁻¹, respectively) in the western Adirondacks than at HF (4.6 kg N ha⁻¹ yr⁻¹) in the central Adirondacks. The greatest increase in nitrate loss in response to the experimental treatments occurred at HF where the HNO₃ additions resulted in the highest nitrate concentrations and lowest N retentions.

Mr. Richard Poirot

Generally, I think the document provides a thorough and useful summary of the state of knowledge relating to the ecological effects of S & N deposition – primarily through acidification and nitrogen enrichment. Several other welfare effects of these pollutants are briefly discussed, although there is no mention of materials damage. As indicated in previous comments, I think it might have also been useful to consider the important welfare effects of sulfate and nitrate aerosols under both this secondary SO_x/NO_x review, as well as in the PM review (copy/paste). Excluding aerosol effects here seems to point to an advance decision that some sort of deposition, critical loads, or critical loads exceedance-type indicators are being considered as a possibility for secondary S and N (hopefully including reduced N) NAAQS.

This flexibility in considering alternative indicators would be a welcome change in the Agency's approach to secondary (or primary) NAAQS, as I think previous reviews have been constrained to consideration of indicators based on the named gaseous criteria pollutants – SO₂ or NO₂ – even though I think the Clean Air Act provides ample encouragement to consider the effects of secondary transformation products and combinations with other pollutants. Since this ISA represents a beginning of the new, streamlined NAAQS review process, its difficult to get a sense of just how far the information and analysis should be taken or “set up” in this document, what belongs in the risk/exposure assessment, and what should be left to the ANPR. If there is really going to be consideration of several deposition-related indicators in this review cycle, I think there should be additional discussion of the options – like critical loads - up front in this ISA, since these are the kind of alternative limits to be evaluated in the risk/exposure assessment, and should definitely not be introduced for the first time in the ANPR.

Another general comment is that I think the document is unnecessarily silent on projected future reductions in S and N emissions that will result from the CAIR rule. We would like to have a better sense of what kinds of effects have occurred, are currently occurring, and are expected to occur as future “on the books” emission reductions occur. At some point before this review is completed, there will likely be some consideration of how NAAQS which relate to location-specific effects and measurements may interact with the receptor location-insensitive emissions cap & trade approach that has become the preferred regulatory approach for further reductions in S and N emissions.

Chapter 2

The 1st half of the chapter's discussion of atmospheric chemistry seems reasonably thorough, although it might more logically follow the discussion of emissions at the beginning of Chapter 3. Its not clear whether this level of detail on atmospheric chemistry is needed in this new-style “Integrated Science Assessment” (as compared to a criteria document or an annex to this ISA). Any excess information here is harmless, but every attempt should be made to make the discussion relevant to those aspects of atmospheric chemistry that may be especially critical to understanding the sources exposures and environmental effects of these S & N pollutants (or other pollutants like Hg the effects of which emissions of these pollutants may affect).

By contrast, the very brief 2-paragraph section 2.5 on Atmospheric Transport conveys almost no useful information (and there's not all that much more detail on transport provided in the Annex). I think this section needs work and might more logically be either tightly focused on those aspects of atmospheric chemistry which in turn affect the transport and ultimate deposition, or expanded and combined with the section on chemical transport models (CTM) at the end of chapter 3.

The 2nd half of chapter 2 seems unnecessarily limited to measurements of the “named” NO_x and SO_x criteria pollutants, with additional tangential discussion of artifactual gains and losses of S & N species in PM mass measurements (which is somewhat beside the point). At the same time, this discussion of measurements, networks, & limitations seems unnecessarily disconnected from discussion of the same topics for measurements, networks & limitations of gaseous & aerosol concentrations, and wet and dry deposition which are included in the first half of Chapter 3 (which actually has relatively little direct discussion of “Ecological Exposures” until the section on Harvard Forest Flux measurements on p. 3-34). I think it would be better to combine all these discussion of measurements of the relevant gaseous, aerosol, wet and dry deposition networks in a single section. Emissions to chemistry & transport to concentrations to deposition and exposures seems like a more logical way to present this information.

Specific Comments

p. 2-1, lines 13-27 (& footnote 1): Possibly you could also include a mention here of the CAA definition of “welfare effects” modified in Section 302 (h) of the 1990 CAA to include the phrase: “...whether caused by transformation, conversion, or combination with other air pollutants”. All gaseous, aerosol and deposition oxidation products of (traditionally defined) SO_x & NO_x emissions should be clearly on the table for consideration, and “combination with other pollutants” may help open the door to considering effects resulting from both oxidized & reduced N emissions.

p. 2-2, line 7: If you changed “particles” to “aerosols” (which includes the suspending gaseous medium), you could leave (gaseous) HNO₃ on the list.

p. 2-3, Figure 2.2-1: Some inorganic nitrates – like NH₄NO₃– can also be transported to remote regions under low temperatures. Generally this section seems overly brief.

p. pages 2-7 to 2-98: This is an interesting and thorough discussion, but there needs to be a clearer connection made between this halogen chemistry and either the emissions, transport or deposition of S & N compounds. You might also add somewhere here that free Cl & Br released by these reactions can also contribute to the conversion of Hg⁰ to Hg²⁺, enhancing the potential for wet & dry Hg deposition to the environment.

p. 2-9 line 29: Add “gaseous” before “species”.

p. 2-16, line 29: Formation of aerosol NH₄NO₃ can also substantially enhance the atmospheric lifetime(s) and transport distance (for both oxidized & reduced N). At some point (not sure where) you should also add some discussion of both current and projected future aerosol

sulfate/nitrate/ammonium/acidity relationships, as lower levels of acidic sulfate may lead to increased levels (& longer transport distances) of aerosol nitrate.

p. 2-16, lines 26-29: This discussion seems overly simplified. Yes, NO and NO₂ are less soluble than SO₂, but they are also much more reactive and, with their “longer-lived reservoir species HNNO₃” do not necessarily transport over longer distances than SO₂ and its transformation products.

p. 2-17, lines 7, 9, 13, 14: Here it looks like you use “NO_x” with several different meanings, and while you claim (line 14) to limit the chapter to FRM/FEM, the chapter quickly moves beyond FRMs for NO/NO₂ to more exotic species & methods.

p. 2-21, lines 15-19: I think this is stated backward. There is more likely to be volatilization loss of NO₃⁻ from (relatively acidic) PM_{2.5} filters (if Teflon – not nylon) and more likely to be positive nitrate artifacts on alkaline PM₁₀ (or PM_{10-2.5} filters), unless preceded by HNO₃ denuders.

p. 2-21, line 22 – p. 2-22, line 8: This is a limited discussion and wimpy illustration of current satellite sensing potential for NO₂. See for example : http://datafedwiki.wustl.edu/index.php/2007-07-18_ESIP_Demo_OMI_NO2 for a better illustration of & links to OMI NO₂ data and images.

p. 2-24, line 22: Traditionally the STN network has employed thermal-optical transmittance (TOT), while the IMPROVE network has used TOR. Currently, STN sites are transitioning to TOR in a phased process – over several years.

p. 2-24, line 23: IMPROVE sample frequency changed from twice a week (Wed & Sat) to 1 in 3 day (same as STN) in Sept. 2000.

p. 2-24, lines 27-31: IMPROVE denuders are coated with a combination of carbonate and glycerin.

p. 2-25, lines 13-22: Its not clear what kind of sampling you’re referring to here. Glass fiber filters with basic coarse particles on the filters would be indicative of TSP samplers currently used primarily for sampling for Pb NAAQS. These are very rarely analyzed for SO₄, NO₃, Cl, or PM mass (which is irrelevant for TSP). Quartz filters can also have (smaller than glass but significant) positive artifacts from SO₂ or HNO₃. Denuders are not really much of an option for hi-vol PM₁₀ samplers – on which quartz filters are typically run, but yes, denuders on lo-vol Teflon filter PM₁₀ or PM_{10-2.5} (by dichot) sampling would help eliminate artifacts.

p. 2-26, lines 6-7: Actually, the loss of pNO₃ from FRM PM_{2.5} samples is likely to be greater than for the “other simple collection systems described above”. Also, the discussion in the following several paragraphs is interesting & informative, but relates primarily to how nitrate sampling losses affect PM_{2.5} mass measurements (not nitrate measurements, which are usually not conducted on these Teflon filters). This reminds me to lament again that its too bad aerosol

nitrate is “off the table” in this NAAQS review – since it is not fully included in our measurement-based definition of PM.

p. 2-27, lines 4,5: I would add “nitrate” after “ammonium” (ammonium sulfate is very stable). Also, changes in the equilibrium between aerosol and gaseous precursors can be due to factors other than T, RH and acidity – such as changes in the precursor gas concentrations - for example rapid loss of (sticky) HNO₃ & NH₃ concentrations in the indoor environment.

p. 2-27, lines 3-21: Why not move the discussion of negative pNH₄ artifacts adjacent to the discussion of positive pNH₄ artifacts beginning line 21, p. 2-25. Also I question your implication that negative artifacts generally exceed positive artifacts for NH₄. Can you provide a reference for this? NH₄ is not measured in IMPROVE and NH₃ denuders are not used in STN – which does attempt to quantify NH₄. What is the intended meaning of the occasional [] in this paragraph?

Chapter 5

This chapter provides a concise summary of the information presented in the preceding chapters. It was generally well-written, but as with the other chapters, there were sections that were choppy (multiple authors?) or where the language would benefit from some more careful editing for clarity. In some cases, the brevity (highly desirable) seems too brief to convey useful information and would benefit from a bit more detail. The use of references is not consistent from section to section and I suggest using more rather than fewer (they don’t take much room and inspire confidence). In some sections the language is unnecessarily vague (“very large”, “higher than might be expected”, etc.) or speculative (“may occur”, “is probably ongoing”, etc.). In many cases, a good point is being introduced that could easily be restated more objectively.

It should be clear that this is a “summary” (which is fine), but not intended as an integrative synthesis or winnowing down of crucial information. There are no new insights provided here, and the chapter doesn’t lead in any obvious directions toward selection of alternative or specific indicators. For the most part, the more detailed and useful information is presented in the preceding chapters. A strong point of Chapter 5 is a series of excellent summary tables at the end (especially Table 5.7.1 on N deposition & effects). These tables start to provide a sense of what kinds of effects are occurring in what regions at what loading rates. I would have liked to see this information taken a bit further to more directly address the concept of critical loads (and exceedances of critical loads), in ways that might provide more advance insights into possible elements (indicators, averaging times, levels & forms) of new secondary NAAQS.

Specific Comments

p. 5-1, line 32: You could add pSO₄ as parallel to pNO₃ in line 29.

p. 5-2, line 7” “controlling SO₂ oxidation” isn’t really the right term here. Could reword as something like “...altering the chemistry of SO₂ oxidation products by neutralizing...”

p. 5-2, line 23: “adequate” (a questionable judgment here), could be improved by adding “for determining compliance with current NAAQS”.

p. 5-3, line 3: “regulatory” doesn’t seem quite right here (& in several other instances). We have no “regulations” for required monitoring of NH₃.

p. 5-3, line 7: You could add a bullet or 2 here describing routine methods/networks and limitations for monitoring of wet (&/or dry) deposition of S & N compounds.

p. 5-3, line 31: Can you be more quantitative than “very little...”

p. 5-4, line 5: Same as previous comment. What % does “substantially smaller” mean?

p. 5-4, line 19: You could add “atmospheric” before “N”.

p. 5-4, line 23: Do you mean “...emitted to the atmosphere...”?

p. 5-4, line 26: There are not “regulatory” monitoring networks for deposition (although maybe there should be).

p. 5-5, lines 23-26: Its not clear what you mean by “riverine flow in the absence of deposition” (some of the riverine flow N is from deposition to upland surface waters or catchments, some is from non-atmospheric sources, and then there’s some directly deposited to the surface waters of the coastal estuary). Also, its not clear what the 11, 5.6 & 5.6 kg N ha⁻¹ refer to here.

p. 5-6, line 21: You could replace “understood” (speculative) with “described in detail”.

p. 5-7, line 23: Can you use a more objective phrase than “thought to be”?

p. 5-8, line 30: Can you use a more objective phrase than “may occur”? For example, have such effects been observed anywhere? If not, you might express these as logical expected outcomes which have not been observed to date...

p. 5-9, line 27: “Consequently” doesn’t seem quite right. Organic matter build-up isn’t necessarily a consequence of “fungi that feed on organic debris...” unless they are less efficient than the bacterial consumers they replace, in which case say so..

p. 5-10, line 1: You could replace “expanded” with something like “reflected a substantial increase in...” The CD didn’t expand knowledge; it just documented it.

p. 5-11, line 16 (& also 5-12, lines 2 and 29 & elsewhere): There are several different indications of a uniquely different situation in the Mid-Appalachian region. It would be useful to provide some explanation of reasons for this difference. Maybe you could do this in the case studies (make it clear there why Shenandoah is different from the Adirondacks) and have pointers to that discussion whenever you indicate a ‘Shenandoah exception’ elsewhere in the document.

p. 5-12, line 28: Add “in” after “decreased”.

p. 5-13, line 2 “Al generally becomes the greater health risk” Greater than what?

p. 5-13, line 19: I assume that by “the most serious effects” you mean something like “more serious than long term”, which I find very counter-intuitive – and would need more convincing to accept as absolute truth. I don’t see any need to play off the severity of chronic vs. acute acidification against each other, nor do I like the policy implication that its only the episodes that matter.

p. 5-15, line 9: This is a good example of overly speculative phrasing (“acidification that has occurred to date has likely been very limited”). You could say roughly the same thing more objectively by indicating that “acidification that has been observed to date has been very limited”).

p. 5-15, line 9,10: Not clear what “Episodic acidification does occur” means here. In the West?

p. 5-15, lines 29-30: I’m surprised that the absolute concentrations of SO_4^{2-} (as well as its % of total ion concentration) did not also decrease. If they did, why not say this as well, as a decrease in the % could be due to an increase in something else.

p. 5-16, line 18 (& next few pages): These case studies seem like they need some additional detail. I’m not quite sure what’s the intended point. Maybe you could put some emphasis on the similarities & differences between the focus regions. For example what were the past & current deposition levels in the Adirondacks – to compare with those reported subsequently for Shenandoah? Possibly an additional ‘case study’ for an acid-sensitive alpine Western ecosystem would round out the regional coverage, and would also help emphasize the extreme sensitivity in these fragile ecosystems which will not benefit from CAIR emissions reductions.

p. 5-16, lines 30-32: This is awkwardly worded. I assume you mean that the number of lakes with $\text{ANC} < 50$ has more than doubled from pre-industrial times (it’s not the estimate of the # of pre-industrial low ANC lakes pre-industrial lakes that’s changed).

p. 5-17, lines 8-10: It seems like these future projections are out of place here. I would move them to after line 23.

p. 5-18, line 32: Add “assessments of” before “the impacts”.

p. 5-21, lines 10, 12-17, 23 & elsewhere: At some point, it would be useful to add some direct discussion of N-fertilized “growth” enhancements vs. other consequences of this fertilization & growth. In some places an impression is given that “growth” is always good, or good up to a point beyond which other adverse consequences can result. Possibly also add some discussion of combined effects of N fertilization + climate change.

p. 5-23, line 4: Can you give a quantitative example of what you mean by “very high”?

p. 5-24, lines 3-4: Can you use a more objective term than “are thought to include”? Could you say something like “Observed effects... have included...”

p 5-24, line 5: Same comment as above. “would be considered relatively low” by whom? Compared to what?

p. 5-27, line 1: This is an interesting observation. I wonder why? Also, “disproportionately high” compared to what?

P 5-27, lines 7-10: It might be helpful to provide some comparative info on N dep rates in Netherlands vs. US.

p. 5-30, line 25 and following paragraph. I think this section should extend beyond the current discussion - focused largely on primary productivity – and describe some of the biogeochemical effects of excess growth and decomposition on dissolved oxygen, etc.

p. 5-30, line 31: Could you say “N-limited”, rather than “N-deficient”?

p. 5-31, lines 18-30: You might add “turbidity” to this list. Its not a “biological” measurement, but neither is DO.

p. 5-33, line 10: Can you state this more objectively than “probably ongoing”?

p. 5-33, line 14: Aren't current inputs in this region much lower than $10 \text{ kg N ha}^{-1} \text{ yr}^{-1}$?

p. 5-34, line 3: You could add “SO₄ deposition and” before “Hg methylation”.

p. 5-34, line 10: Why are DIC & DOC in ()?

p. 5-36, line 19: There are no “regulatory networks” for S or N deposition.

p. 5-36, line 20: You could add “, elevational gradients and” before “hotspots” to emphasize a particularly important inadequacy of existing deposition networks.

p. 5-36, line 22: Add a (cite) where indicated.

p. 5-37, lines 2-4: Is it possible you meant “high elevation areas of the West” in line 4? Otherwise including both the Appalachians and high elevation seems redundant. And if you did mean the “West” (which would be true) you might describe the first region as “New York / New England” to include the Adirondacks & Catskills. Possibly also add a mention of sensitive areas in Canada which might benefit from future reductions in US S and or N emissions.

p. 5-37, line 11: I hesitate to criticize this presentation of quantitative “limits” here, since I think more such numbers would be useful in this section, but in this case, I think N deposition rates are already below this $9\text{-}15 \text{ kg N ha}^{-1} \text{ yr}^{-1}$ limit, while the low bound limit of $0 \text{ kg S ha}^{-1} \text{ yr}^{-1}$ suggests that no amount of S reduction would prevent re-acidification (which doesn't make sense). It might also be useful here to provide some indications of how these ranges compare to both current deposition levels and future projections under of CAIR and contrast these with the estimated damage thresholds in both Adirondacks and Shenandoah.

p. 5-37, line 14: As in a previous comment, you are implying here that “growth” in some ecosystems is a beneficial outcome – to be contrasted with “adverse effects” in others. I think this could be fixed by changing “ecosystems” to something like “ecosystem components”.

p. 5-37, line 19: Insert “Adding” before “excess” or change “to” to “in”.

p. 5-37, line 26: Should be “West” coast.

p. 5-39, end of 1st line under Nitrate: Add s to 1980. And 2nd to last line: Could you add example parenthetical “X ueq/L)” to indicate what is meant by “Very High”.

p. 5-42: Table 5.7.1 provides a very useful quantitative summary of effects of N deposition on acidification and nutrient enrichment that have been identified in recent studies. Would it be possible to provide a similar summary table providing quantitative of S deposition (or S+N deposition where applicable) on acidification? It is curious that these concluding summary tables include some international observations but seem to have no Canadian references.

Dr. Armistead Russell

While I think the current ISA for Oxides of Nitrogen and Sulfur-Environmental Criteria provides much of the needed information for the process of reviewing the secondary NAAQS for NO_x and SO_x, at present the document is in need of significant modification to provide such information efficiently and effectively. The document should be read and edited for relevancy of the information being provided, and how it is provided, to the ultimate goal of the review of, and providing the scientific basis for possibly changing, the associated secondary NAAQS.

One step in this process would be to look at Chapter 5 and edit what is presented there to specifically provide a summary of what information is needed to provide a scientific foundation for a review of the standard. Having edited the summary chapter, then go back and write the chapters to bring forth the specific points needed to provide the basis for what is in Chapter 5.

Unlike the ISA's for the two associated primary NAAQS, this document begins in Chapter 2 with the more traditional Criteria Document approach of reviewing the atmospheric physics and chemistry of NO_x and SO_x, as opposed to a "Source to Dose" presentation, which here would be source to exposure or load. The latter approach led to a much more efficient presentation of the key concepts, data and science. As it currently reads, aspects of Chapter 3 seemed to be somewhat repetitive of information in Chapter 2, and the flow is awkward. Taking a source to exposure/load also leads back to the key idea that the increased environmental exposures can be linked to a source, which is what is controlled. Maybe it is due, in part, to lack of scientific investigation, but it is this type of information that is required for this review. In revising the current Draft ISA, it might be good to have someone read Chapter 5, the Conclusions, first, and then identify what are the major points, and what is needed to support the key findings as to the environmental implications of anthropogenically-influenced increased exposures and loads. Then, the degree to which those key findings are supported in Chapters 1-4 can be assessed, and the discussion of tangential information can be streamlined.

Chapter 2:

As noted above, I think the ISA would benefit from a Chapter 2 that goes from Sources to Environmental Exposure/Load. Chapter 3 should be integrated in. In addition to having atmospheric chemistry and physics, aquatic and soil chemistry and physics should be added. This would save much repetition later on in the report (e.g., about what leads to acidification, its affect, mobilization of aluminum, etc.). Fig. 2.2-1 could have a twin(s) showing what is happening in surface water and the soil-water matrix.

On page 2-3, line 16, one should note they are referring to US emissions. Lines 19-23 are a bit confusing at present. What is important, here, is that virtually all of the fuel-bound sulfur gets oxidized to a volatile component (SO₂ or SO₃), and that there is very little natural, oxidized sulfur in air over the US, so the sulfur emitted from burning a fuel is quantitatively related to that in the fuel.

The section on Measurement Methods needs to be refocused. The major question to be addressed here is if the current methods employed in the field provide reliable measurements of NO_x , NH_3 , NH_4^+ , NO_3^- , SO_2 and SO_4^{2-} for levels of interest, and this should be answered quantitatively. At present, there is discussion of the various measurement approaches (some is needed) and lots of discussion on possible interferences, but never does one get the answer to what is the typical uncertainty in the measurements at a typical monitor in the US and how that impacts our ability to accurately quantify environmental exposures. I suspect that the methods employed for most of the species, while subject to some interferences, provides fine data, and that the level of uncertainty is such that we need not concern ourselves with possible interferences and biases. The major exception, of course, is ammonia, which is not even discussed. What really limits our ability to quantify deposition accurately (e.g., do we need more accurate measurements, different kinds?). Quantify the problems, let the reader assess if they are of concern.

More specific comments:

2-1:24-27: What about pSO_4 ?

Rxn 2.2-2: Balance the reaction.

2-5:13: Reactions 2.2-1 and 2.2-2.

Section 2.2.2 It is not apparent the need for so much on Cl chemistry. Also, this section should be preceded by a more comprehensive discussion of N_2O_5 . I would be very tempted to minimize this discussion.

2-9:25: Not sure why you use monomeric. What polymers do you have in mind?

2-10: 28: In both cases, NO_3 and H are radicals, not ions.

2-11:2 Ravishankara.

2-13:28: A second reaction for comparison should be added (i.e., the ozone reaction).

2-14:10: What chemistry of NO_3^- formation is being discussed?

2-15:1: Ammonium nitrate formation as well.

2-17:30 Remove "all".

2-21: Section 2.6.2.2 needs to address accuracy more directly.

2-22...: Section 2.6.3 should be more quantitative.

A section on NH_3 measurement is needed.

2-27: Section 2.7.1: It is weird that pNO_3 is included here, pSO_4 is not.

2-28:28: There is a summary paragraph for NH_3 that is about as long as those for the other species, but where is this drawn from?

At present, the summary is a bit sparse.

Chapter 3

The Chapter begins with a discussion of sources, which as noted above, should go in a more integrated Chapter 2. It seems a bit unnatural to not start “the story” with sources. Also, while this chapter starts with sources, one does not get a quantification of such. One should not have to go to the Annexes to get a reasonably quantitative overview. A single table showing the contributions of, say, mobile, non-mobile, utility, biogenic and agricultural sources to oxidized NO_x, N₂O, reduced N and SO_x is required. This might be done on a tons of S and N basis for ready comparison between the oxidized and reduced forms. Some estimate of the transport to North America might also be provided. (A sister Table showing the fate of the above is also suggested.)

A Table of fate of reduced N, oxidized N and SO_x is suggested, showing the masses dry and wet deposited by species, as well as the amount transported away from the continent and associated estuaries. Having this table integrated with the table of sources might be of interest from a mass balance perspective.

Figure 3.7-4 As discussed during the SO_x-Primary ISA review, this figure is not very effective. First it refers to “... in focus” in focus of what? Further, most of the data is very much at the bottom end, so it is difficult to see what is really happening, and it needs to include more information (what years...). I would have it provide the mean, median, 5 & 95%iles, and the extreme value. Further, it might be given on a log scale as most of the lower level information is now lost.

There is a bit of a contradiction in Chapter 3. On page 3-8, line 11, it says the ratio of N-to-S is 14, close to that in the tissue. The prior line says that approximately half of the S is in the ash. Please explain further.

In considering emissions, it would be good to also provide some information as to future emissions for perspective. CAIR is going to significantly lower emissions in areas where they are currently high. This is important for our further consideration as to how a standard might impact air quality.

Section 3.13 Summary.

This section starts out very roughly. 3.13.1 reads more like a set of bullet points in sentence form. The last paragraph (3-161, 14-6): Is this what you really mean to say, and from what part of the report is it distilled? Table 3.11.1 is rather more concrete at 21%, but does not indicate the reduced N fraction. Also, the first sentence of 3.12.2 is not really taken from something in the text (I might add this to the prior text).

3-60:26 change to “...surface of some coastal...”

What this chapter (or what should become a section of Chapter 2) needs is to focus on what one wants to know, as this section currently has some stuff I like to know, and some of what is not needed to assess a NAAQS. What is really needed? I would say a best estimate (map) of total N

and S deposition, a list of estuaries suffering eutrophication with estimates (and uncertainties) of what is contributing. There was too little on ammonia(um) deposition.

Specific comments:

3-2:30: This sentence contains a non-sequiter.

3-3:2: Reactions, not reactivities.

3-4:16: I would remove “small”

3-4:24: “... for example, **is estimated to have** decreased...”

3-9:16: Remove “However”

3-13:13 “is a smaller **loss process** than its net thermal decomposition”

3-13:28 Remove “which”

3-14: Section 3.5.5. the source strength given is not a source strength unless one knows the volume. Further, more information on how robust this value might be nationally is needed.

3-23: Section 3.7 is somewhat repetitive of prior sections.

3-23:23-29: What is this paragraph trying to say?

3-23:28: Higher than what?

3-24:1-8: Much more quantitative information is available, e.g., annual average concentrations of the various individual oxidized nitrogen species.

Figure 3.7-2: Put Figure 3.7-3 right below this figure for comparison. Also, can you provide the same figures for oxidized nitrogen species of interest?

3-26:2 “for more complete descriptions”

3-26:7: This is not seen in Fig. 3.7-4.

3-28:6: “correlative”?

3-28:29 “...mean **oxidized** N...”

3-36:6: Units on deposition velocity?

3-36:7-7: Is this a major extrapolation. Please clarify.

Fig. 3.9-3: How significant is “a”, and the units for concentration are non-standard.

3-41:3-5: Further explanation as to “why” is needed.

3-43:26-27: Same should be added for regional scale.

3-47:19-17: I would note all of the evaluation work and measurements done by/for the state, RPO’s and EPA. It may not get published in the open literature, but these groups probably do most of the model evaluations done.

3-48:12 “This”?

3-50: What is needed here are the best estimates of N deposition using CMAQ.

Figure 3.12-3: How about adding NH₃?

Chapter 4:

Chapter 4 reads more like the old Criteria Documents, being rather exhaustive and sometimes appearing to lose the point.

Also, maybe I missed this, but this chapter seems to have ad hoc decided to use SO₄⁺ instead of using SO₄⁼ (or SO₄²⁻). Why?

There is significant repetition in this chapter. Much of what is in 4.2.1.4 is in pages 4.10-12.

4-1:22: interacting**ng**
4-2:6: "...when **the** nutrient..."
Figure .1-1: For the forest critical chemical limit, what are the units of 1.0? For the Lake, is "0" sensible?
4-7:13 Might you add surface water acidification?
4-8:4: Define Oa
4-8-29: What about NH_x?
Fig. 4.2-1: Wrong figure. (Switch with 4.2-2)
4-11:25: Replace "This" with "The"
4-13:22: "... forested areas, it is ..."
4-15:27 Define "%" of what.
4-16: "...acidic deposition mobilizes..." no comma.
4-16:14-16: Repetitive.
4-19:11-12: Same paragraph.
4-19:16: "...general... generally..."
4-19:28 "or neutralizing"
Fig. 4.2-3 Probably should define ANC in the caption and also add the 0 lines.
4-22:30 All the "in summary" statements get a bit old.
4-24:3: [K]
Section 4.2.22: Repetitive of prior material.
Fig. 4.2-6: "Bad death"? As opposed to "Good death"?
4-39:25-26: Sentence needs a bit more explanation.
4-43:19: "linear**ly**"
4-44:31: Context needed for 5.0, 5.5 and 6.0. Reference to what effects.
4-49: Should not ANC measurement be up front in measurement methods?
4-49:9: Repetitive.
4-49:14: Small is not defined, and why is this viewed as small?
4-49:23-24: Unclear clause at the end.
4-50: Lines 21-22 and 25-26 appear to be contradictory.
4-54:20: Be specific as to at what level.
4-64:23: Lawrence no longer in review, I think.
4-123:L24...: Again, seems repetitive.
4-129:17 Reviewing only the negative effects appears one-sided.
4-130:2: "Results of these studies..."
4-132:30: Confirmed? Support?
4-135:1: What would this equate to in terms of deposition?
4-148:29: define stges.
4-150:26: "probably"? Cite.
4-152.19-27: Largely repetitive.
4-154:5-9 The term significant here must be purely definitional. What is meant by, or determines, significant?
4-156:4...: Repetitive.
4-157:14 "Niwt Ridge where..." remove comma.
Section 4.4.1.2 can be shortened.
4-168:19-21. This need not be stated. It has to be.

- 4-174:3-14. Be quantitative... at what levels are the impacts found?
- 4-177:12-31 Use same units as before.
- 4-177:3-: Provide units on coefficients.
- 4-178:9-14: Discuss exactly what was measured, provide uncertainties.
- 4-179:24-25: Is this likely an issue?
- 4-179: Provide a summary.

Chapter 5:

This chapter is still a bit choppy, but primarily should be revised by asking the question: What information specifically is needed to provide the scientific basis for reviewing the NAAQS secondary NO_x and SO_x standards.

Specifics:

- 5-3:8-12: What about ammonia?
- 5-12:28: "... decreased **in** some..."
- 5-12:31: be a bit more specific as to how.
- 5-13:29-30: Could just be my memory, but I don't recall this.
- 5-16-28: Remove extra "."
- 5-16:30 Try not to use "many" as opposed to giving a specific fraction.
- 5-18:5: rearrange sentence" "contributions of CA...acidity) from some ..."
- 5-20:16: "...section discusses..."
- 5-20:31: "is it "of" or "by"?"
- 5-26:8-28: These are important points, but they did not seem to come out very distinctly in the chapters. (same with 5-27:15-20).
- 5-28:22: "AQCD, **further studies suggest** productivity..."
- 5-31:6: awkward sentence.
- 5-37:16 remove "in"

Overall: Probably the single biggest limitation in this document is acknowledged early on: It does not try to establish critical loads, without which it becomes difficult to know what levels of environmental exposures are acceptable or not. Decision makers and advisors need such information, even if it is fraught with "uncertainties." (I put uncertainties in quotation marks as it seems as though we use that as a reason to not go forward, which is not the case.) The second real need of this ISA is to dig more in to the reduced nitrogen deposition issue.

Mr. David Shaw

Chapter 1 Comments

Message of the document

This chapter drives the whole document and as such it should say up front that a major part of this assessment is examining. A primary unit of measure is shown throughout as deposition, so it must state the strength of a *deposition* approach to both of these contaminants. I also feel that the purpose and consequences of evaluating N and S in tandem should be addressed. While the chapter opens with a statement of eliminating particulates in the scope of this scientific analysis, I do not feel that this is not an appropriate message.

Framing Questions

Some of the framing questions (pg 1-2) are awkward (e.g. bullet 3 line 13) and don't focus the work as well as the more succinct language below (proposed by E.Cowling). Individual Comments on the September 2007 Draft Plan for Review of the Secondary National Air Quality Standards for Nitrogen Dioxide and Sulfur Dioxide October 26, 2007. I suggest substituting the framing questions for these:

“What scientific evidence and/or scientific insights have been developed since the last review to indicate if the current public-health based and/or the current public-welfare based NAAQS need to be revised or if alternative levels, indicators, statistical forms, or averaging times of these standards are needed to protect public health with an adequate margin of safety and to protect public welfare?”

“What scientific evidence and/or scientific insights have been developed since the last review to indicate whether, and if so, what particular ecosystem components or other air-quality-related public welfare values, are more or less sensitive than the populations of humans for which primary standards are established and for this reason may require a different level, indicator, statistical form, or averaging time of a secondary standard in order to protect public welfare?”

Format

The format calls for a summary of findings at the end of each major segment of each chapter which the authors do, but they should also, as much as possible, discuss the relevance of each of those findings with the two questions above. This should be expressed in Section 1.3. The format should also require that the conclusions in Chapter 5 restate these original questions and summarize all findings within the two reworded framing questions. The summaries would serve the document best by ensuring that all the material is summarized in a clear and concise manner.

The deposition monitoring network maps are too small to show adequacy of the network.

Gaps in legislative background and history of the current review

CAIR should be mentioned in the regulation portion of this discussion.

Major scientific, multi-state and national surveys and summary reports of the period (i.e., since SO₂ NAAQS 1982) need to be mentioned, included but not limited to the EPA Eastern and Western Lake Surveys, the National Acid Precipitation Assessment Program (NAPAP), the NAPAP Integrated Assessment in 1991, and the EPA Acid Deposition Standard Feasibility Study Report to Congress in 1995. Significant state surveys like the Adirondack Lakes Survey in 1984-1987 and the ongoing Adirondack Long Term Monitoring of lakes and streams should also be included.

The recent progression of the importance of N in acidification should be given more attention. For example, the recent chemical recovery signal in Adirondack lakes appears closely linked with N deposition. Recent data show stream acidification is also highly correlated.

Section 1.1.3 SO_x. Include the primary standard for SO_x. Fill in the history gap between 1988 and 2006.

Section 1.1.4. Include what led to the development of a deposition approach to this secondary standard as opposed to an ambient air standard. Add what other states may have done with respect to 1) more stringent primary standards or 2) proposals of secondary standards or other regulations to protect the environment from NO_x and SO_x.

Current levels of ambient and deposition N and S are improved because of all efforts to attain clean air, which should include major state efforts such as New York State's NYS Sulfur Dioxide Control Program in 1985 that evolved into its Acid Deposition Control Program. Several northeast states (NH, VT, CT and ME) have discussed or proposed S and N deposition standards to protect forests.

Particulate of NO_x and SO_x omission

The first paragraph states "The scope of the joint NO_x and SO_x ISA is limited to welfare topics that do not duplicate those addressed by the forthcoming particulate matter (PM) science assessment. The welfare effects of visibility impairment and climate interactions associated with particulate NO_x and SO_x will be addressed within the secondary PM NAAQS." The omission of particulate phase NO_x and SO_x limits the potential for setting standards in the future using both PM and oxides of Nitrogen and Sulfur standards. Furthermore, PM plays a significant role in nitrogen and sulfur deposition. I feel that separating out the effects of gas- versus aerosol-phase S/N will be difficult, since wet and dry deposition can include both phases, and atmospheric chemistry and transport affect both phases. The ISA clearly states that "particulate NO_x and SO_x will be addressed with the secondary PM NAAQS review," and it therefore becomes crucial that these two review process tracks are highly consistent with each other. One cannot proceed independently of the other track.

Implementation of a secondary standard(s)

It is important to state that this document covers the setting of a secondary standard(s) but not, at this point, the implementation of a secondary NAAQS. They would need to be developed or This would be very difficult because at present, there are no adequate source-receptor models to provide the level of control that would be required to meet the secondary NAAQS, if the deposition standards were exceeded in any particular area.

Charge Questions

Question #1

To what extent is the evidence on atmospheric chemistry and physics, air quality, and deposition and exposure sufficiently and correctly described, clearly communicated, and relevant to the review of the secondary NAAQS for NO₂ and SO₂?

- The information on gas-phase chemistry and physics is sufficient and well-presented in Chapters 2 and 3.
- Since both gas-phase and particulate S/N are involved in deposition, the ISA would benefit from inclusion of more chemistry and physics on the particulate side in these chapters as well.
- Clearly communicated – see “Format” section. Summaries are graded as poor.

Question #2

How well characterized are the relevant properties of the ambient air concentrations and deposition of NO_x and SO_x, including policy-relevant background concentrations, spatial and temporal patterns, and the relationships between ambient air concentrations and ecological exposures?

- There is enough information on urban NO₂ and SO₂ to characterize ambient levels, but not enough information in rural areas and sensitive ecosystems.
- In general, emission inventories for SO₂, whose sources are well-characterized, are more reliable than estimates for emissions of NO_x, and certainly far more reliable than NH₃ emissions.
- There is more information, in terms of spatial coverage, on wet deposition of SO₄, NO₃, and NH₄ than ambient SO₂, NO₂, or NH₃

Question #3

How sufficient is the information on atmospheric sciences and exposures for the purposes of evaluating and interpreting the ecological effects presented in Chapter 4 of the draft ISA?

There is an inherent danger here of assuming that all areas of ecological effects are equally important or equally developed in the literature, which in my view, they are not. The danger is

the inevitable conclusion that the sufficiency of the information will not be adequate for all of them.

A better use of time would be to go to the bigger questions framed above that have to do with the interaction between the primary and the secondary standards, and the value and need for establishing deposition standards.

General Comments

Deposition-based Standard

The draft ISA demonstrates a strong interrelationship between deposition data and effects to ecosystems as shown in Table 5.7-1. Concentration-based standards are not biologically relevant to most resources at risk from air pollution (surface waters, groundwater, soils, etc). It is important that other EPA programs (i.e. CAMD) are reviewing or participating in this ISA. All programs should be aware of the proposed move forward to a multi-pollutant approach and this new pathway.

One or multiple secondary standard

One secondary standard for the US or several according to various sensitive receptor areas in the country? Primary standards protect only one organism, people, and they are distributed throughout the country, so generally one size does fit all. Secondary standards protect sensitive ecological receptor areas (ER) which are not evenly distributed and have multiple sensitive components within them. Are we setting one secondary standard for the country? Have we identified all of the sensitive environments that we wish to track and to protect?

Within each sensitive environment, have the ecological assessments identified a single or multiple deposition targets? Shouldn't each ecological area have one targeted? Have the major eutrophication-stressed estuaries picked a limit for NO_x and NH_x?

Critical loads may be an area of interest. I feel there is a need for discussion on the state of knowledge of critical loads. While there may not be a high resolution understanding (small blocks) areas can be defined by forest or east vs west. The EU has critical loads in place.

Emphasis on Certainty

While there is uncertainty surrounding some data or modeling, I feel that it is important to emphasize that solid and certain data does exist which the scientific community has accepted.

Climate change

There may be a need for analysis of the effects of the SO_x and NO_x deposition on climate change. In addition, the effects of climate change on SO_x and NO_x pollution.

Adequacy of existing air monitoring network

- The density of SO₂ and NO₂ monitors is adequate to track the primary standard, but is not adequate for the secondary standard.
- The bulk of the ambient SO₂ and NO₂ monitors, as well as the Speciation Trend Network for fine particulate SO₄ and NO₃, are located in urban and suburban areas.

- There is no national NH₃ monitoring program, and what little information is available for ambient concentrations is research-oriented.
- There is information on fine particulate SO₄ and NO₃ at >100 Class I areas through the IMPROVE program, but this is probably not adequate to characterize all sensitive ecosystems across the nation.
- The major deposition networks (CASTNet, NADP/NTN) were generally established to track the effects of emissions reductions from the energy generation sector.
- In terms of data coverage, there are more wet deposition monitors measuring SO₄, NO₃, and NH₄ than there are ambient NO₂ monitors, however these networks are not generally capable of characterizing deposition in urban areas or many rural areas (especially in the western US).
- Dry deposition of S/N is inferred rather than measured directly by routine networks like CASTNet, but even so there are far fewer dry deposition sites compared to wet.

Transformation Products

Clarify which N pollutants are being discussed. Research which oxidized N compounds are relevant and feasible to monitor. While NO_y may be more appropriate, there is a need to discuss the difficulty of measuring individual compounds. Chapter 2 does describe some of the heterogeneous chemistry between NO_y and halogens, but does not really cover ammonium nitrate/sulfate formation. Halogen chemistry plays an important role in urban coastal settings, but ammonium nitrate/sulfate chemistry is important over vast regions of the continent.

NO_x as a precursor

NO_x should be discussed as a precursor to the following:

- Ozone
- Ambient nitrates
- N deposition
- Fine particulates

Inclusion of N₂O

N₂O is certainly a greenhouse gas, but is not a component of NO_y, nor is it relevant to O₃/PM/haze formation or N deposition (and subsequent acidification or nutrient enrichment). More information on particulate N would be favored over N₂O analysis.

Dry deposition

There are different monitoring networks for sulfur and nitrogen. Monitoring network not adequate for dry deposition

Emissions inventories

Are emission inventories adequate for each, SO_x, NO_x and NH_x? For the primary and secondary standard? In general, emission inventories for SO₂, whose sources are pretty well-characterized, are more reliable than estimates for emissions of NO_x, and certainly far more reliable than NH₃ emissions.

Multiple indicators and multiple sensitive areas.

States may contain several sensitive regions. Sensitive regions like the Adirondack Mountains will have multiple natural resources with different indicators and/or critical thresholds (see Figure 4.1.-1, page 4.4). Have all the sensitive areas within the US been identified (Alaska and Hawaii have none?)? Will each state be responsible for verifying these areas? Is there is enough data to do that in each area? Or whether there will be a blanket single indicator for each sensitive region? What is the more quantifiable indicator of ecological health? Has the effects literature (for the case study areas where the literature is more complete) identified the most representative indicator to protect the whole area?

Other editorial comments:

1. Figure 4.2-2 wrong diagram for the caption.
2. pg 4-41, line 6 Figure 4.2-2 should be Figure 4.2-8.
3. pg 5-10 line 12. Spring chemistry is not baseflow condition for streams in the Eastern US.
4. Glossary. Add base cation surplus, chronic acidification, episodic acidification...

Dr. Kathleen Weathers

This chapter was rather unbalanced in writing style as well as analysis and synthesis. It was often difficult to identify the salient points (bottom lines) in much of the first part of the document (until 4.3). There were inaccuracies and often unqualified (under referenced), value-laden language was used (see specific comments, below). Consider reorganizing and rewriting the chapter, using one voice, and organizing the chapter around a conceptual model. The integrative tables that are at the end of the document could be used to guide a discussion on the overarching effects of acidification and nitrogen enrichment on ecosystems. Supporting information could then be revised, condensed and used to underpin this synthesis.

4a How well are the major effects of SO_x and NO_x on ecological acidification identified and characterized?

4b To what extent do the discussion and the integration of evidence across scales (e.g., species, communities and ecosystems and regions) correctly represent and clearly communicate the state of the science?

The major acidification effects, which have been indirectly or directly linked to the (wet, sometimes wet + dry) deposition of sulfur and nitrogen, have been identified and characterized in this chapter. The relative importance and certainty of various effects was less well characterized.

Categorization of biological and chemical effects is a useful way to partition.

Again, I suggest adding a conceptual model to the beginning of this section. The table (identified as Fig. 4.1-1) and its associated description is a start. However, I do think that it is more logical to consider deposition as a driving variable and various ecosystem (i.e., export rates, nutrient or pollutant cycling, acidification process, productivity) or ecological properties as response to variables rather than the other way around, despite the suggestion that working from top to bottom is inherent in the critical load approach.

Much of the text details studies that have explored the effects of acidification on the various foci of ecological studies (as listed above). The coverage of the literature is not as broad as it might be; it relies heavily on a limited set of research/researchers' work. There is also much supporting literature in this chapter from past decades. It is very useful to cite these foundational studies, but also important to note what the relevant (to this time period) bottom lines are, especially in situations where the environmental conditions have changed significantly in the interim. One suggestion is to identify what is new (recent results, or synthesis efforts) and what has been known for some time. I am concerned that studies whose results, for example, point to how ecosystems responded to the depositional regime in the 1980s and 1990s may be dismissed, or questioned as relevant in 2008. However, there have been a number of integrated studies over the past several years, both modeling efforts and synthesis whose results are cited. Thus, it would be useful to identify which class of results is timeless, i.e., where the findings still hold despite significant changes in emissions and/or deposition over time. Integration and synthesis is a challenging task.

5. How well has the ISI characterized the relationship between acidifying deposition levels of NO_x and SO_x and ecological effects.

The characterization could be made more direct. As one example, studies that highlight deposition, whether estimated across the landscape or to a site (e.g., Lovett and Reuth 1999), or controlled through experimental additions (Harvard Forest, Catskills, Bear Brook additions, for example), were not highlighted or cited well throughout the text.

The ecosystem approach was underscored in the first part of the chapter. Thus framing the relationships of deposition and ecological responses using a mass balance approach would be another logical way to organize the discussion. In this way, it would be possible to move from the influence of S and N on watersheds, to communities and species within watersheds. Consider including a table or a section that summarizes the relationships between deposition and ecological effects toward the beginning of the chapter.

I appreciated the introduction of critical loads as a way that the EU and others have considered successfully the relationship between atmospheric deposition and environmental effects; it is an important approach to explore in this document.

The word/concept “thresholds” is used in numerous cases. It should be (re) defined in this section. A summary section or table identifying ecological thresholds that have been documented (the response variable and the actual threshold) would be very useful. Even if it turns out that few have been defined, it would be useful exercise.

An introductory paragraph about the (legitimate/defensible) use of models and a brief characterization (i.e., table about the nature and utility of the various models, and their results, that are cited throughout this chapter) would be a useful addition. Also, wherever there are multiple model results for an ecosystem or region, a summary of the similarities (e.g., page 4-87 for the Adirondacks) as well as the differences in model results and whether there has been any independent validation or verification of the models should accompany the comparison. It is also critical to point out which models consider biological interactions (the bio part of biogeochemistry) and which do not. Since N is very biologically active, and tree species can have a considerable effect on N loss from ecosystems (e.g., Lovett et al. 2000, 2002, 2004), use of any models that do not consider the influence of biology on geochemical cycles should be very well justified. And, the details, strengths and weaknesses of models that do include biological processing should likewise be revealed.

6. How well characterized are the oxidized and reduced forms of nitrogen on acidification

The N cascade section was good. Consider including the first part of the N cascade section and a parallel sulfur section at beginning of this chapter.

Make clear that in addition to HNO₃ wet and dry deposition, some areas of the country are experiencing a rise in NH₃ emissions and NH₄ wet deposition (NADP data show this increase). Although there are fundamental differences in ecological processing when reduced vs oxidized

nitrogen is added to an ecosystem, it would be useful to point out that there may be downstream acidification/eutrophication effects if nitrogen demand is exceeded and nitrogen is leached as a result of NH₄-N deposition (in addition to the brief mention on page 4-145).

A few overarching comments:

Total deposition, wet+snow+dry+fog, in places where it matters, is still poorly characterized for most places on the planet, especially in non-homogeneous terrain (e.g., see Weathers et. al. 2000, 2006). Getting reasonable estimates of total deposition, especially across space, and relating emissions to deposition are still active areas of research. These facts can be problematic when deposition is used as the independent variable or treatment for any of a suite of ecological responses. I do not think that this challenge — obtaining and using reasonable estimates of deposition — renders specific studies either inadequate or wrong. However, it does underscore the need for multiple lines of evidence to identify, with some confidence, the effects of N and S on loading ecosystems, species, and communities. These multiple lines of evidence often come through the use of experimentation (especially loading studies), modeling, theory, long-term analyses, and use of deposition gradients (where there are surely large differences in deposition even if the absolute numbers have large uncertainties associated with them). I think that pointing out the importance and power of multiple lines of evidence in supporting current understanding about the effects of NO_x and SO_x on ecological systems would be a useful thing to do at the beginning of this section.

I like the case studies, but they could be used more effectively, perhaps to highlight parts of a conceptual model. Make clear why particular case studies are being used.

Some details:

The charge on sulfate is incorrectly identified throughout the text: sulfate is an anion, not a cation, and it's divalent, not monovalent. I note, however, that there is correct inference for sulfate's behavior throughout the text (i.e. it is listed/included in discussions about mobile anions).

Decomposition should be added as an important process in section 4.1.1 and food webs vs food chains are more realistic descriptions of energy movement within ecosystems.

There are many places in the text where references are missing, but necessary. The references also seem quite biased toward specific researchers or research groups.

Correct the text for reduction/oxidation reactions and products (the transformation from ammonium to nitrate is oxidation, for example).

Figures 4.2-1 and 4.2-2 are transposed.

Y axis on Fig. 4.2-6: "Bad?" death?

Do the colors mean anything on Fig. 4.2-12?

It seems to me that much of the fish data that include abundance, biodiversity, and total numbers of species present as response variables are potentially confounded by freshwater stocking activities. Perhaps stocking did not influence the results or was not practiced in many of the focal surface waters. If that is true, it would help to say so, if it is not, it may still be worth mentioning this confounding point.

I believe that the original reference to N saturation was Agren and Bosatta (1988) (page 4-106).

Equating throughfall N flux with atmospheric N deposition is not warranted in many locations in the US because of variable uptake and leaching in the canopy (page 4-110). Throughfall deposition or flux is often considered to be a good indicator of what is deposited to the forest floor, but not necessarily what is deposited from the atmosphere to canopies when the nutrient, ion or element of interest is biologically reactive/active. Sulfur, however, has often been considered an exception. In regard to sulfur in throughfall, research that has suggested its utility as an indicator of total deposition should be included as well as the one on page 4-162 (Cape) that questions it. For example, many papers, including Garten and Lindberg; Lindberg and Johnson; Lovett; Weathers et al.; and more have pointed to its utility.

Words such as “harmful” or “negative effects” without qualification should be avoided; if used they should be qualified with specific environmental terms (e.g., harmful physiological effects of acidity on fish biota). Also “ecosystem or ecological health” is ill defined and often a lightning rod-word. I suggest defining it, or better yet, don’t use it.

Consider doing a publication count by year on the topic of acidification/critical loads.

Listed below are some relevant articles whose results should be considered when revising the ISA:

- Butler, T. J., G. E. Likens, F. M. Vermeylen, and B. J. B. Stunder. 2005. The impact of changing nitrogen oxide emissions on wet and dry nitrogen deposition in the northeastern USA. *Atmos. Environ.* 39:4851-4862.
- Burgin, A.J. and S. K. Hamilton. 2007. Have we overemphasized the role of denitrification in aquatic ecosystems? A review of nitrate removal pathways. *Front. Ecol. Environ.* 5:89-96 [indirect effects of S on N biogeochemistry]
- Caraco, N.F. 1993. Disturbance of the phosphorous cycle: a case of indirect effects of human activity. *Trends in Evolution and Ecology* 8:51-55. [indirect effects of S on P biogeochemistry]
- Caraco, N.F., J.J. Cole, G.E. Likens, G.M. Lovett, and K.C. Weathers. 2003. Variation in NO₃ export from flowing waters of vastly different sizes: Does one model fit all? *Ecosystems* 6:344-352. [N deposition and what influences nitrate in aquatic systems draining various source areas]
- Groffman, P.M., J.S. Baron, T. Blett, A.J. Gold, I. Goodman, L.H. Gunderson, B. Levinson, M. Palmer, H.W. Paerl, G.D. Peterson, N.L. Poff, D.W. Rejeski, J.F. Reynolds, M.G. Turner, K.C. Weathers and J. Wiens. 2006. Ecological thresholds: An important concept with no practical application, or the key to successful environmental management? *Ecosystems* 9:1-13. [a very cursory examination of the concept and difficulties surrounding thresholds; acid deposition used as an example]
- Kelly, V.R., G.M. Lovett, K.C. Weathers, and G.E. Likens. 2005. Trends in atmospheric ammonium concentrations in relation to atmospheric sulfate and local agriculture. *Environmental Pollution* 135:363-369.

- Kelly, V.R., G.M. Lovett, K.C. Weathers, and G.E. Likens. 2002. Trends in atmospheric concentration and deposition compared to regional and local pollutant emissions at a rural site in southeastern New York, USA. *Atmospheric Environment* 36:1569-1575.
- Likens, G. E., D. C. Buso, and T. J. Butler. 2005. Long-term relationships between SO₂ and NO_x emissions and SO₄²⁻ and NO₃⁻ concentration in bulk deposition at the Hubbard Brook Experimental Forest, New Hampshire. *J. Environ. Monitor.* 7(10):964-968.
- Lovett, G.M., K.C. Weathers, and M.A. Arthur. 2002. Control of nitrogen loss from forested watersheds by soil C:N ratio and tree species composition. *Ecosystems* 5:712-718.
- Lovett, G.M., K.C. Weathers, and M.A. Arthur. 2004. The influence of tree species on nitrogen cycling in the Catskill Mountains, New York. *Biogeochemistry* 67:289-308.
- Lovett, G.M., K.C. Weathers, and M.A. Arthur. 2001. Is nitrate in stream water an indicator of forest ecosystem health in the Catskills. Pp. 23-30. *In: M.S. Adams (ed.). Catskill Ecosystem Health.* Purple Mountain Press, Fleischmanns, New York.
- Lovett, G.M. 1994. Atmospheric deposition of nutrients and pollutants to north America: an ecological perspective. *Ecol. Apps.* 4:629-650.
- Lovett, G.M. and H. Reuth. 1999. Soil nitrogen transformations in beech and maple stands along a nitrogen deposition gradient. *Ecol. Apps.* 9:1330-1334.
- McNeil, B.E., J.M. Read and C.T. Driscoll. 2007. Foliar nitrogen responses to elevated atmospheric nitrogen deposition in nine temperate forest canopy species. *Environ. Sci. and Technol* 41:5191-5197.
- Simkin, S.M., D.N. Lewis, K.C. Weathers, G.M. Lovett, and K. Schwarz. 2004. Determination of sulfate, nitrate and chloride in throughfall using ion-exchange resins. *Water, Air and Soil Pollution* 153: 343-354.
- Templer, P.H., M.A. Arthur, G.M. Lovett and K.C. Weathers. 2007. Plant and Soil Natural Abundance ¹⁵N: Indicators of relative rates of nitrogen cycling in temperate forest ecosystems. *Oecologia*. 153: 399-406.
- Templer, P.H., G.M. Lovett, K.C. Weathers, S.E.G. Findlay, and T.E. Dawson. 2005. Influence of tree species on ¹⁵N sinks and forest N retention in the Catskill Mountains, New York, USA. *Ecosystems* 8:1-16.
- Weathers, K.C., G.E. Likens, and T.J. Butler. 2006. Acid rain. Pp. pp. 1507-1522. *In: W. Rom (ed.). Environmental and Occupational Medicine, 4th edition.* Lippincott-Raven and Wilkins Publishers, Philadelphia.
- Weathers, K.C., S.M. Simkin, G.M. Lovett, and S.E. Lindberg. 2006. Empirical modeling of atmospheric deposition in mountainous landscapes. *Ecological Applications* 16:1590-1607. [Review of deposition in heterogeneous terrain, use of throughfall sulfate as an indicator of total deposition; quantification of hotspots of S and N deposition, in general and Acadia and Great Smoky Mountain National Parks, in particular]
- Weathers, K.C., G.M. Lovett, G.E. Likens, and R. Lathrop. 2000. The effect of landscape features on deposition to Hunter Mountain, Catskill Mountains, New York. *Ecological Applications* 10:528-540. [quantification of hotspots of S and N deposition]
- Weathers, K.C., G.E. Likens, F.H. Bormann, J.S. Eaton, W.B. Bowden, J.L. Anderson, D.A. Cass, J.N. Galloway, W.C. Keene, K.D. Kimball, P. Huth, and D. Smiley. 1986. A regional acidic cloud/fog event in the eastern United States. *Nature* 319:657-658. [importance of fog or clouds in delivering acidifying substances to ecosystems].