

**Summary Minutes of the U.S. EPA Clean Air Scientific Advisory Committee (CASAC)  
NO<sub>x</sub> and SO<sub>x</sub> Secondary NAAQS Review Panel  
Public Meeting on April 2-3, 2008**

Panel Members: See Panel Roster provided in Attachment A.

Date and Time: April 2 – 3, 2008

Location: Marriott at Research Triangle Park  
4700 Guardian Drive, Durham, NC

Purpose: To conduct a peer review of EPA's *Integrated Science Assessment for NO<sub>x</sub> and SO<sub>x</sub> – Environmental Criteria* (First External Review Draft) and a consultation on the EPA's *Draft Scope and Methods Plan for Risk/Exposure Assessment: Secondary NAAQS Review for NO<sub>x</sub> and SO<sub>x</sub>* (March 2008). Both of these documents may be found at <http://www.epa.gov/ttn/naaq/standards/no2so2sec/index.html> .

Attendees:

|                |                                                                                                                                                                                                                                                                           |
|----------------|---------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|
| Chair:         | Dr. Armistead (Ted) Russell                                                                                                                                                                                                                                               |
| CASAC Members: | Dr. Ellis B. Cowling<br>Dr. Douglas Crawford-Brown (by phone)<br>Dr. Donna Kenski                                                                                                                                                                                         |
| Panel Members: | Dr. Praveen Amar<br>Dr. Andrzej Bytnerowicz<br>Ms. Lauraine Chestnut<br>Dr. Paul J. Hanson<br>Dr. Rudolf Husar<br>Dr. Dale Johnson (by phone)<br>Dr. Naresh Kumar<br>Dr. Myron Mitchell<br>Mr. Richard L. Poirot<br>Mr. David J. Shaw (by phone)<br>Dr. Kathleen Weathers |
| EPA SAB Staff: | Ms. Kyndall Barry, Designated Federal Officer<br>Dr. Anthony Maciorowski, Deputy Director                                                                                                                                                                                 |

Other EPA Staff: Jeffrey Arnold, EPA  
Allen C. Basala, EPA  
Tim Benner, EPA  
Ila Cote, EPA  
Jean-Jacques Dubois, EPA  
Tara Greaver, EPA  
Dave Guinnup, EPA  
Jeffrey Herrick, EPA  
Bryan Hubbell, EPA  
Lingli Liu, EPA  
Tom Long, EPA  
Ellen Lorang, EPA  
Jason Lynch, EPA  
Karen Martin, EPA  
Sarah Mazur, EPA  
Kristopher Novak, EPA  
Anne Rea, EPA  
Mary Ross, EPA  
Vicki Sandiford, EPA  
Ginger Tennant, EPA  
Paul Wagner, EPA  
Randy Waite, EPA  
Debra Walsh, EPA  
Nealson Watkins, EPA  
Lydia Wegman, EPA

Other Participants: Jamie Cajka, RTI  
Bill Cooter, RTI  
Jack Cosby, University of Virginia  
Marion Deerhake, RTI  
Jon M. Heuss, Alliance of Automobile Manufacturers (by phone)  
John Jansen, Southern Company  
Cindy Langworthy, Hunton & Williams  
Ona Papageorgiou, NYSDEC  
Liz Schoeneck, ICFI  
Anthony R. White, American Heart Association (AHA)  
Kate Winston, Inside EPA

Attachments: (A) NO<sub>x</sub> & SO<sub>x</sub> Secondary Review Panel roster; (B) agenda; (C) Federal Register notice announcing the meeting; (D) “Background and History” presentation by OAR and ORD; (E) “Highlights of the Draft ISA” presentation by ORD; (F) “Highlights of the Draft SM” presentation by OAR; (G) preliminary ISA comments by the Panel; (H) preliminary SM comments by the Panel; and (I) public comments.

## Meeting Summary

The discussion followed the issues and general timing as presented in the meeting agenda (Attachment B).

### **Wednesday, April 2, 2008**

Ms. Kyndall Barry convened the meeting and explained that the CASAC NO<sub>x</sub> & SO<sub>x</sub> Secondary Review Panel will operate under the Federal Advisory Committee Act (FACA). She also announced that there would be a conference call on May 5, 2008, for the CASAC to review and, possibly, approve the Panel's letter to the EPA Administrator concerning the draft ISA. Dr. Anthony Maciorowski thanked the Panel for their hard work. He also thanked staff members from EPA and members of the public for attending the meeting.

Dr. Armistead (Ted) Russell, thanked the EPA staff for preparing the draft ISA and the draft Scope and Methods Plan. The Panel was introduced, and Dr. Russell then reviewed the agenda. He noted that the Panel would draft the ISA letter by the end of the day, and would vote on the main points of draft letter the following day.

Dr. Ila Cote of EPA's Office of Research and Development (ORD) welcomed everyone and introduced Dr. Karen Martin from EPA's Office of Air and Radiation (OAR). Dr. Martin presented *Background and History of the Review of the Secondary NO<sub>2</sub> and SO<sub>2</sub> NAAQS* (Attachment D). During the presentation, she emphasized the court-ordered deadlines, summarized the history of the previous reviews of the welfare effects of NO<sub>2</sub> and SO<sub>2</sub>, and described the Agency's approach to standard-setting for the NAAQS criteria pollutants. Dr. Martin also noted the Agency has decided to separate the visibility effects of NO<sub>x</sub> and SO<sub>x</sub> (from the current review) for inclusion in the PM NAAQS review.

Drs. Mary Ross, Jeffrey Arnold, Tara Greaver, and Paul F. Wagner, EPA ORD, presented *Integrated Science Assessment for NO<sub>x</sub> and SO<sub>x</sub> – Environmental Criteria* (Attachment E). The presentation highlighted the elements contained in the ISA and included the Agency's charge questions to the Panel. Major points from the Agency's presentation: the inclusion of reduced forms of nitrogen (NH<sub>x</sub>) in the review; sources of NO<sub>x</sub>, NH<sub>x</sub>, and SO<sub>2</sub> emissions; average annual deposition of inorganic N and total S; chronic and episodic acidification; regional variability and appropriate case studies; various indicators of terrestrial and freshwater nitrogen enrichment; and categorization of methylmercury (MeHg) and greenhouse gases as other welfare effects. The Panel commended the Agency on the inclusion of NH<sub>x</sub> in the current review. Panelists engaged the ORD presenters on the definition of adverse effects in the ISA and the need for better characterization of deposition, acidification, and enrichment.

During the public comment period, Mr. Jon M. Heuss provided comments on behalf of the Alliance of Automobile Manufacturers (Attachment I). Mr. Heuss noted that his comments focused on the science that will influence the NO<sub>x</sub> secondary standard. He cited data that indicated there has been a decrease in the past decade in the vehicular contribution to NO<sub>x</sub> and SO<sub>x</sub> levels due to improvements in control technologies. Mr. Heuss closed with the point that

discussions of further decreases in emissions should be considered in the context of reductions already achieved.

Following the public comment period, the discussion moved to Agency charge questions 1 – 3 (Attachment B1). Panelists presented their various comments captured in *preliminary ISA comments* (Attachment G). The Panel stressed the following ways to improve the ISA: contracting a third-party editor to improve the transitions in the document; more detail on  $\text{NH}_x$  emissions and inclusion of total reactive nitrogen,  $\text{N}_r$ ; relocating the information on the monitoring networks from the Annexes to Chapter 2; addressing existing data gaps which may require EPA to include other monitoring networks with publicly available data; adding a qualitative discussion of the models (CMAQ, GEOS, CASTNET, etc) being used; establishment of the links from emissions to concentration to deposition; adding greater coverage of European studies; and including diagrams of  $\text{NO}_x$  and  $\text{NH}_x$  emissions.

After a short break, the Panel addressed charge questions 4 – 6. Panelists again urged ORD to focus on the links between emissions, deposition, loads, and effects to improve the organization and context of the document. Major discussion points included: the use of indirect and direct effects to balance evidence; inclusion of nitrogen's role in increasing carbon sequestration as a beneficial ecological effect; the need to amplify discussions of nutrient depletion and subsequent recovery of ecosystems; and the relationship between incidents of windthrow and forest fires and increased nitrogen emissions. Dialogue on the appropriate indicators of acidification focused on the sugar maple, red spruce, dogwood species; although grasslands and aspen were also mentioned. The Panel also discussed the use of terrestrial and aquatic mobile anions to unify the sulfur and nitrogen components of Chapter 4.

Following the lunch break, the Panel addressed the remainder of the charge questions starting with charge questions 7 – 9. Dr. Ellis Cowling, the CASAC representative on SAB Integrated Nitrogen Committee (INC), presented the INC's 31 October 2007 resolution (see Dr. Cowling's comments in Attachment G). While the effects of  $\text{NO}_x$  on nitrogen enrichment were well characterized, the Panel urged the Agency to move to an integrated approach and better characterize the contribution of oxidized and reduced forms of nitrogen. Desert ecosystems, herbaceous plants and shrubs, and microbials were identified for inclusion in the case studies. Other recommendations included: figures with deposition plotted against the response variable of choice; a qualitative discussion of the dose-response relationship where data was not available; the need to address other factors contributing to nitrogen loss (e.g. forest composition); and the need to move from use of "beneficial" and "negative" when describing effects. Due to the overlap in the work of the INC's and the  $\text{NO}_x$  and  $\text{SO}_x$  Secondary Panel, Dr. Maciorowski provided the following caveat on the sharing of information between advisory panels: because the INC's report is still a work-in-progress, it cannot be considered advice until it is finalized and vetted by the Science Advisory Board in compliance with FACA regulations.

In response to charge question 10, the Panel pointed out the following omissions from the ISA chapter on "other welfare effects": a statement of the effects included and omitted from the review and/or primary outcomes of the document; a reference to the PM review for discussion of the effects of  $\text{NO}_x$  and  $\text{SO}_x$  on visibility; the interaction of nitrogen on carbon sequestration; and the direct effect of nitric acid on vegetation and dry deposition. Panelists engaged the EPA on

the handling of MeHg and greenhouse gasses: recommending that the linkages between mercury and sulfur and N<sub>2</sub>O, a green house gas, should be included in the document, but de-emphasized. Additionally, the suggestion to de-emphasize PAN (page 4-176) was made because it is not a significant contributor to nitrogen deposition: though toxic, it is episodic and primarily confined to urban areas.

Discussions continued on to the final charge questions, 11 and 12. The Panel unanimously agreed that Chapter 5 was well-written and concise, and proposed that it be brought forward as an executive summary of the ISA. Additional recommendations included: the expansion of the summary tables in the executive summary and conclusion; strengthening the qualitative conclusions with numerical data; and adding economic analyses to the case studies. To improve the transition from the technical information presented in the ISA to the forthcoming risk and exposure assessments, the Panel suggested the solidifying the economic valuation of the effects and/or economic services impacted. The summary tables start to link effects and indicators, and the incorporation of the economic analysis into the case studies (e.g., Adirondacks and Chesapeake Bay) should proceed next.

At the end of the day, Dr. Russell reiterated the letter will address each individual question, tasked the Panel members to compose response to their respective charge questions, and summarized the major review comments discussed by the CASAC Panel. Ms. Barry adjourned the meeting for the day.

#### **Thursday, April 3, 2008**

Ms. Barry reconvened the meeting of the CASAC NO<sub>x</sub> and SO<sub>x</sub> Secondary NAAQS Review Panel. During the public comment period, Ms. Ona Papageorgiou, representative of NY State Department of Environmental Conservation (NYSDEC), suggested that EPA take a different approach in the Scope & Methods document. She noted that EPA uses statements like “depending on available data” and “if feasible”, and asked EPA to specifically identify where the monitors would need to be physically located to fill these data gaps. She also noted that references to the existing data needed to be included and particulate emissions should be included in the comprehensive discussion of welfare effects of NO<sub>x</sub> and SO<sub>x</sub>.

Following the public comment period, the Panel discussed the draft responses to the ISA charge questions. Each paragraph was projected onto the screen and discussed by the Panel. By the end of the session, the draft letter to the EPA Administrator was outlined and consensus reached on the major points as required by FACA. Ms. Barry then laid out the next steps in the letter-approval process: revisions to the working draft due on April 18<sup>th</sup>, the draft letter will be posted on the SAB Staff Office Web site by April 25<sup>th</sup>, and the final review and approval by the statutory CASAC will take place via teleconference on May 5<sup>th</sup>. Dr. Russell asked EPA if they had any final questions for the Panel. Dr. Ross thanked everyone for their comments and suggestions.

After the morning break; Drs. Dave Guinnup, Anne Rea, Bryan Hubbell, Mr. Randy Waite, and Ms. Lydia Wegman, of EPA’s Office of Air and Radiation (OAR) presented *Overview of the Scope and Methods Plan Supporting the Review of the Secondary NO<sub>2</sub> /SO<sub>2</sub> NAAQS* (Attachment

F). The presentation summarized the Agency's seven-step approach for the risk and exposure assessments (REA) to go from exposure to ecological effect (e.g., ecosystem effects, relevant biological exposure indicators, and ecosystem responses, services and valuation). EPA identified the welfare effects central to the REA: aquatic acidification, terrestrial acidification, aquatic nutrient enrichment, and terrestrial nutrient enrichment, with qualitative discussions of MeHg and N<sub>2</sub>O.

Discussions between the Panel and OAR turned to selection of atmospheric indicators for the welfare standards and the need for linkages to the science. Effects based on total loadings must be translated to deposition and, finally, to ambient air concentrations for rulemaking. The Panel engaged EPA on the feasibility of considering alternative indicators, including N<sub>r</sub>, as well as oxidized and reduced forms of nitrogen. EPA noted that new indicators could be considered, but the criteria pollutant for regulatory purposes would remain listed as NO<sub>x</sub>.

The Panel then moved to address the Agency charge questions 1 – 7 (Attachment B2). While the Panel commended the Agency on the scope and structure of the REA, they offered the following suggestions: strengthening the use of indicators and indices to more clearly define the relationship; a preference to use actual monitoring data augmented with model results; use of “decrease” instead of “reduce” because “reduce” is ambiguous and refers to a chemical reaction; use of existing regional models to assist the Agency's prioritization of case studies; and public availability of any data utilized for the REA.

Following the lunch break, the Panel responded to the remainder of the charge questions. Panelists urged the Agency to consider model predictions of critical load, which has proved successful for Canada and European nations. It was also noted that the US Forest Service is developing a national-scale, critical load model, although it is currently only conceptual. Other areas of concern raised included: the challenge to resolve the divergent scales of models and data; a proposed approach to define effects in terms of economic valuation; and the need to include the role of N<sub>r</sub> in carbon sequestration.

At the close of the meeting, the Panel was reminded of the April 18<sup>th</sup> deadline for revisions to the draft ISA letter and the follow-on CASAC teleconference to approve the draft letter on May 5, 2008. After noting that the final comments on the Scope and Methods document were due by April 25, 2008, Ms. Barry adjourned the meeting.

Respectfully Submitted:

/Signed/

Ms. Kyndall Barry  
Designated Federal Officer

Certified as True:

/Signed/

Dr. Ted Russell, Chair  
CASAC NO<sub>x</sub> & SO<sub>x</sub> Secondary  
NAAQS Review Panel

**NOTE AND DISCLAIMER:** The minutes of this public meeting reflect diverse ideas and suggestions offered by committee members during the course of deliberations within the meeting. Such ideas, suggestions, and deliberations do not necessarily reflect definitive consensus advice from the panel members. The reader is cautioned to not rely on the minutes to represent final, approved, consensus advice and recommendations offered to the Agency. Such advice and recommendations may be found in the final advisories, commentaries, letters, or reports prepared and transmitted to the EPA Administrator following the public meetings.

Attachment A: Roster of CASAC NO<sub>x</sub> & SO<sub>x</sub> Secondary NAAQS Review Panel

**U.S. Environmental Protection Agency  
Clean Air Scientific Advisory Committee  
NO<sub>x</sub> & SO<sub>x</sub> 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**

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**U.S. Environmental Protection Agency  
Clean Air Scientific Advisory Committee (CASAC)  
NO<sub>x</sub> & SO<sub>x</sub> Secondary NAAQS Review Panel  
Public Meeting: April 2-3, 2008**

**Marriott at Research Triangle Park, 4700 Guardian Drive, Durham, NC, 27703**

**Purpose:** To conduct a peer review of EPA's Integrated Science Assessment (ISA) for Oxides of Nitrogen and Sulfur--Environmental Criteria (First External Review Draft) (EPA/600/R-07/145, December 2007) and a consultation on the EPA's draft Scope and Methods Plan for Risk/Exposure Assessment: Secondary NAAQS Review for Oxides of Nitrogen and Oxides of Sulfur (March 2008 Draft).

**Wednesday, 2 April 2008**

|            |                                                                     |                                                                                                                                                       |
|------------|---------------------------------------------------------------------|-------------------------------------------------------------------------------------------------------------------------------------------------------|
| 8:30 a.m.  | Welcome                                                             | Ms. Kyndall Barry, EPA SAB Staff Office, Designated Federal Officer<br><br>Dr. Anthony Maciorowski, Deputy Director, EPA SAB Staff Office             |
| 8:40 a.m.  | Introduction of Members, Review Agenda                              | Dr. Ted Russell, Chair                                                                                                                                |
| 8:50 a.m.  | Background and History of the NAAQS                                 | Dr. Ila Cote<br>EPA's Office of Research and Development (ORD)<br><br>Ms. Lydia Wegman<br>Dr. Karen Martin<br>EPA's Office of Air and Radiation (OAR) |
| 9:05 a.m.  | Highlights of Draft ISA and Agency Charge Questions (Attachment B1) | Dr. Jeffrey R. Arnold<br>Dr. Tara Greaver<br>Dr. Paul F. Wagner<br><br>EPA ORD                                                                        |
| 9:35 a.m.  | Public Comment Period                                               | To be announced                                                                                                                                       |
| 9:45 a.m.  | Response to Charge Questions 1 – 3                                  | <u>Dr. Praveen Amar</u><br>Dr. Rudolf Husar<br>Dr. Naresh Kumar<br>Dr. Donna Kenski<br>Dr. David Shaw                                                 |
| 11:00 a.m. | Break                                                               |                                                                                                                                                       |
| 11:15 a.m. | Response to Charge Questions 4 – 6                                  | <u>Dr. Dale Johnson (by phone)</u><br>Dr. Kathleen Weathers                                                                                           |
| 12:15 p.m. | Lunch                                                               |                                                                                                                                                       |

|           |                                      |                                                                               |
|-----------|--------------------------------------|-------------------------------------------------------------------------------|
| 1:30 p.m. | Charge Questions 7 – 9               | <u>Dr. Ellis Cowling</u><br>Dr. Andrzej Bytnerowicz                           |
| 3:00 p.m. | Break                                |                                                                               |
| 3:15 p.m. | Response to Charge Question 10       | <u>Dr. Paul Hanson</u><br>Dr. Myron Mitchell                                  |
| 3:45 p.m. | Response to Charge Questions 11 – 12 | <u>Dr. Douglas Crawford-Brown</u><br>Ms. Lauraine Chestnut<br>Mr. Rich Poirot |
| 4:45 p.m. | Summary of Major Review Comments     | Dr. Ted Russell                                                               |
| 5:30 p.m. | Adjourn Meeting                      | Ms. Kyndall Barry                                                             |

**Thursday, 3 April 2008**

|            |                                                                                                |                                                                                                           |
|------------|------------------------------------------------------------------------------------------------|-----------------------------------------------------------------------------------------------------------|
| 8:30 a.m.  | Reconvene the Panel Meeting                                                                    | Ms. Kyndall Barry                                                                                         |
| 8:35 a.m.  | Public Comment Period                                                                          | To be announced                                                                                           |
| 8:50 a.m.  | Discussion of Draft Responses to ISA Charge Questions                                          | Dr. Russell and Panel                                                                                     |
| 10:15 a.m. | Break                                                                                          |                                                                                                           |
| 10:30 a.m. | Highlights of the Draft Scope and Methods Document and Agency Charge Questions (Attachment B2) | Dr. Dave Guinnup<br>Dr. Anne Rea<br>EPA OAR                                                               |
| 11:00 a.m. | Response to Charge Questions 1 – 2                                                             | <u>Dr. Douglas Crawford-Brown</u><br>Dr. Paul Hanson<br>Dr. Rudolf Husar<br>Dr. Naresh Kumar              |
| 11:30 p.m. | Response to Charge Questions 3 – 7                                                             | <u>Dr. Ellis Cowling</u><br>Dr. Myron Mitchell<br>Dr. David Shaw                                          |
| 12:15 p.m. | Lunch                                                                                          |                                                                                                           |
| 1:00 p.m.  | Charge Questions 3 – 7 continued                                                               |                                                                                                           |
| 1:30 p.m.  | Response to Charge Questions 8 – 10                                                            | <u>Mr. Rich Poirot</u><br>Dr. Andrzej Bytnerowicz<br>Dr. Dale Johnson (by phone)<br>Dr. Kathleen Weathers |
| 2:30 p.m.  | Response to Charge Question 11                                                                 | <u>Dr. Donna Kenski</u><br>Dr. Praveen Amar<br>Ms. Lauraine Chestnut                                      |
| 3:00 p.m.  | Summary and Next Steps                                                                         | Dr. Ted Russell                                                                                           |
| 3:30 p.m.  | Adjournment                                                                                    | Ms. Kyndall Barry                                                                                         |

## Attachment B1: Agency ISA Charge Questions

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 NO<sub>2</sub> and SO<sub>2</sub>?
2. How well characterized are the relevant properties of the ambient air concentrations and deposition of NO<sub>x</sub> and SO<sub>x</sub>, 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<sub>x</sub> and SO<sub>x</sub> 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<sub>x</sub> and SO<sub>x</sub> and environmental effects?
6. How well characterized is the relative importance of the oxidized and the reduced forms of nitrogen on ecosystem acidification?
7. How well are the major effects of NO<sub>x</sub> 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?
10. Several additional effects are discussed, including mercury methylation, direct gas-phase effects on foliage, and N<sub>2</sub>O as a greenhouse gas. How well does the draft ISA characterize the evidence on these topics?
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?

## Attachment B2: Agency Scope & Methods Plan Charge Questions

1. In outlining the scope of this risk/exposure assessment, we have created a flow diagram that represents how nitrogen and sulfur compounds move from 'source to dose' in the environment (see Figure 2-1). How adequately does this conceptual model for evaluating risks due to deposition-related ecological effects characterize what should be covered in the scope of this assessment?
2. The main ecosystem effects areas we anticipate evaluating in this risk/exposure assessment are (1) risks to terrestrial ecosystems from nitrogen enrichment effects, (2) risks to aquatic ecosystems from nitrogen enrichment effects (eutrophication), (3) risks to terrestrial ecosystems from acidification effects (nitrogen and sulfur), and (4) risks to aquatic ecosystems from acidification effects (nitrogen and sulfur). We also plan to qualitatively discuss the role of sulfur enrichment on methylmercury production and the role of nitrous oxide in climate change. What key effects areas, if any, have been overlooked by this approach? Should the assessment plan be modified to include other effects?
3. Due to the complexity of conducting a nationwide risk/exposure assessment for welfare effects due to NO<sub>x</sub> and SO<sub>x</sub>, we have outlined a strategy designed to identify sensitive ecosystems and a range of harmful/adverse effects (see Figure 3-1). The seven steps are to (1) identify documented biological, chemical and ecological effects and potential ecosystem services, (2) define sensitive areas using GIS mapping, (3) select risk/exposure case study assessment areas, (4) evaluate current loads and effects in case study assessment areas, (5) scale up the case study assessment areas to larger sensitive areas where feasible, (6) assess current ecological conditions in those areas, and (7) assess alternative levels of protection under different ambient scenarios. Does the Panel agree with this general approach? Should it be improved or modified?
4. In the seven-step approach to the current conditions risk/exposure assessment, Step 1 (Section 3.1) describes an approach to identify the documented effects, biological, chemical and ecological indicators, and potential ecosystem services related to acidification and nutrient enrichment. Does the Panel agree with this approach or can they suggest alternative approaches we should consider?
5. In the seven-step approach to the current conditions risk/exposure assessment, Step 2 (Section 3.2) outlines a path to define areas sensitive to total reactive nitrogen and sulfur inputs. Do the Panel members agree with this approach or are there better alternatives that should be considered?
  - a. We are attempting to characterize the risks to ecosystems from sulfur and nitrogen deposition nationwide by clustering sensitive ecosystems where possible and by using the linkages between these areas at different scales. Please comment on the adequacy of this approach.
  - b. How appropriate are the datasets and GIS maps listed in Table 3-4 for identifying ecosystems sensitive to nitrogen and sulfur and/or are there others that have been overlooked?
6. In the seven-step approach to the current conditions risk/exposure assessment, Step 3 (Section 3.3) outlines a path to identifying risk/exposure case study assessment areas.
  - a. Table 3-5 provides an initial list of indicators, mapping layers and multimedia models that may be used to assess ecosystem risk and exposure. Please comment on the appropriateness of these and suggest alternatives that may be better suited for this analysis.
  - b. Please comment on the list of potential case study assessment areas in Table 3-6 and Table 3-7 and make recommendations or suggest any alternatives.

7. In the seven-step approach to the current conditions risk/exposure assessment, Step 4 (Section 3.4) outlines a path to assess current nitrogen and sulfur loads and their effects on case study assessment areas. Does the Panel agree with how we have described our approach to identifying datasets, gaps, and uncertainties?
  - a. We have initially identified the primary chemical indicator that is most suitable for assessing ecosystem acidification effects as acid neutralizing capacity (ANC), with alternatives depending on data availability (see section 3.4.1 and Appendix B). Does the Panel agree with this selection, or can they suggest alternative/additional key indicators?
  - b. We have described the models being considered for this analysis (see section 3.4.2 and Appendix C). Does the Panel agree with the choice of these models, and can they help prioritize them for modeling the responses of the indicators recommended in Step 1 (Section 3.1)?
8. In the seven-step approach to the current conditions risk/exposure assessment, Step 5 (Section 3.5) discusses how to scale up case study areas to more spatially extensive sensitive areas, where appropriate. Does the Panel agree with this approach or can they suggest alternatives?
9. In the seven-step approach to the current conditions risk/exposure assessment, Step 6 (Section 3.6) outlines a path to assess the current conditions of sensitive ecosystems. How well does the Panel agree with the approach outlined for calculating response curves and utilizing mapping and ecosystem services to characterize current conditions or can the Panel recommend alternative approaches?
  - a. How well does the Panel agree with using ecosystem services to provide a common metric for comparing ecological risks due to nitrogen and sulfur deposition effects?
  - b. How well does the Panel agree with collecting current valuation studies to understand the value of bundled ecosystem services? Can the Panel recommend additional or alternative approaches?
10. In the seven-step approach to the current conditions risk/exposure assessment, Step 7 (Section 3.7) describes an approach to assess degrees of protection/levels of effects under alternative forms and levels of ambient NO<sub>x</sub> and SO<sub>x</sub> standards. This approach attempts to describe how the methods, models, and results of the current conditions risk/exposure assessment can inform our evaluation of the appropriate form(s) and level(s) of a national standard. How well does the Panel agree with the approach outlined in this section, the issues presented, and the 9 steps outlined to assess potential forms and levels of the standard? Please suggest any additional or alternative steps we should take into consideration.
11. Additional ecological/welfare effects due to NO<sub>x</sub> and SO<sub>x</sub> emissions that we do not currently anticipate evaluating in detail in this review include the following:
  - Nitrogen saturation,
  - Maple decline,
  - Ammonia air deposition and toxicity to native mussels,
  - Relationships between acidity/nutrient enrichment and mercury methylation,
  - Sensitive areas for acidity/nutrient enrichment impacts, identified from biogeochemical characteristics, and
  - Climate change effects due to N<sub>2</sub>O.

Does the Panel agree that these represent lower priority effects for the current assessment? If not, what does the Panel recommend?

Attachment C: Federal Register Notice announcing the meeting

[Federal Register: February 26, 2008 (Volume 73, Number 38)]  
[Notices]  
[Page 10243-10244]  
From the Federal Register Online via GPO Access [wais.access.gpo.gov]  
[DOCID:fr26fe08-48]

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ENVIRONMENTAL PROTECTION AGENCY  
[FRL-8533-5]

Science Advisory Board Staff Office; Clean Air Scientific  
Advisory Committee (CASAC); Notification of a Public Advisory Committee  
Meeting and Teleconference of the CASAC Oxides of Nitrogen (NO<sub>x</sub>) &  
Sulfur Oxides (SO<sub>x</sub>) Secondary NAAQS Review Panel

AGENCY: Environmental Protection Agency (EPA).  
ACTION: Notice.

-----  
SUMMARY: The Environmental Protection Agency (EPA) Science Advisory Board  
(SAB) Staff Office announces a public meeting of the Clean Air Scientific  
Advisory Committee Oxides of Nitrogen (NO<sub>x</sub>) and Sulfur Oxides (SO<sub>x</sub>) Secondary  
National Ambient Air Quality Standards (NAAQS) Review Panel (CASAC Panel) and  
a public teleconference of the chartered CASAC. The CASAC Panel will conduct  
a peer review of EPA's Draft Integrated Science Assessment for Oxides of  
Nitrogen and Sulfur--Environmental Criteria (First External Review Draft)  
(EPA/600/R-07/145, December 2007) and a consultation on the EPA's draft Scope  
and Methods Plan for Risk/Exposure Assessment: Secondary NAAQS Review for  
Oxides of Nitrogen and Oxides of Sulfur. The chartered CASAC will review and  
approve the Panel's report by public teleconference.

DATES: The CASAC Panel will meet from 8:30 a.m. on Wednesday, April 2, 2008  
through 4 p.m. Thursday, April 3, 2008 (Eastern Time). The chartered CASAC  
will meet by public teleconference at 10 a.m. on Monday, May 5, 2008 (Eastern  
Time).

ADDRESSES: The April 2-3, 2008 public meeting, will take place at the  
Marriott at Research Triangle Park, 4700 Guardian Drive, Durham, NC 27703,  
telephone: (919) 941-6200. The May 5, 2008 public teleconference, will be  
conducted by phone only.

FOR FURTHER INFORMATION CONTACT: Any member of the public who wants further  
information concerning the April 2-3, 2008 meeting, may contact Ms. Kyndall  
Barry, Designated Federal Officer (DFO), EPA Science Advisory Board (1400F),  
U.S. Environmental Protection Agency, 1200 Pennsylvania Avenue, NW.,  
Washington, DC 20460; via telephone/voice mail: (202) 343-9868; fax: (202)  
233-0643; or e-mail at: [barry.kyndall@epa.gov](mailto:barry.kyndall@epa.gov). For information on the CASAC  
teleconference on May 5, 2008, please contact Mr. Fred Butterfield,  
Designated Federal Officer (DFO), at the above listed address; via  
telephone/voice mail: (202) 343-9994 or e-mail at: [butterfield.fred@epa.gov](mailto:butterfield.fred@epa.gov).  
General information concerning the CASAC can be found on the EPA Web site at:  
<http://yosemite.epa.gov/sab/sabpeople.nsf/WebCommittees/CASAC>.

SUPPLEMENTARY INFORMATION:

Background: The Clean Air Scientific Advisory Committee (CASAC) was  
established under section 109(d)(2) of the Clean Air Act (CAA or Act) (42  
U.S.C. 7409) as an independent scientific advisory committee. CASAC provides  
advice, information and recommendations on the scientific and technical  
aspects of air quality criteria and national ambient air quality standards

(NAAQS) under sections 108 and 109 of the Act. The CASAC is a Federal advisory committee chartered under the Federal Advisory Committee Act (FACA), as amended, 5 U.S.C., App. The Panel will comply with the provisions of FACA and all appropriate SAB Staff Office procedural policies.

Section 109(d)(1) of the CAA requires that the Agency periodically review and revise, as appropriate, the air quality criteria and the NAAQS for the six ``criteria'' air pollutants, including NOX and SOX. EPA published the Integrated Review Plan for the Secondary National Ambient Air Quality Standards for Nitrogen Dioxide and Sulfur Dioxide (Final) in December 2007. The CASAC Panel provided a consultation on the draft Plan in October 2007: ([http://yosemite.epa.gov/sab/sabproduct.nsf/77B813F50BDD96C1852573A70005BAF3/\\$File/casac-08-003.pdf](http://yosemite.epa.gov/sab/sabproduct.nsf/77B813F50BDD96C1852573A70005BAF3/$File/casac-08-003.pdf)). EPA's Office of Research and Development (ORD) has completed the Draft Integrated Science Assessment for Oxides of Nitrogen and Sulfur--Environmental Criteria (ISA) and EPA's Office of Air and Radiation (OAR) will also release a Scope and Methods Plan for Risk/Exposure Assessment. The purpose of the April 2-3, 2008 meeting,

[[Page 10244]]

is for the CASAC Panel to provide advice on these two documents. The chartered CASAC will meet by conference call to review and approve the Panel's draft report on the ISA.

Technical Contacts: Any questions concerning EPA's Draft Integrated Science Assessment for Oxides of Nitrogen and Sulfur--Environmental Criteria (First External Review Draft) should be directed to Dr. Tara Greaver, ORD, at (919) 541-2435 or [greaver.tara@epa.gov](mailto:greaver.tara@epa.gov). Any questions concerning EPA's Scope and Methods Plan for Risk/Exposure Assessment: Secondary NAAQS Review for Oxides of Nitrogen and Oxides of Sulfur should be directed to Dr. Anne Rea, OAR, at (919) 541-0053 or [rea.anna@epa.gov](mailto:rea.anna@epa.gov).

Availability of Meeting Materials: EPA-ORD's Draft Integrated Science Assessment for Oxides of Nitrogen and Sulfur--Environmental Criteria (First External Review Draft) can be accessed at [http://www.epa.gov/ttn/naaqs/standards/no2so2sec/cr\\_pd.html](http://www.epa.gov/ttn/naaqs/standards/no2so2sec/cr_pd.html). EPA-OAR's Scope and Methods Plan for Risk/Exposure Assessment: Secondary NAAQS Review for Oxides of Nitrogen and Oxides of Sulfur will be accessible at [http://www.epa.gov/ttn/naaqs/standards/no2so2sec/cr\\_pd.html](http://www.epa.gov/ttn/naaqs/standards/no2so2sec/cr_pd.html). The agenda and other materials for this CASAC teleconference will be posted on the SAB Web site at: <http://www.epa.gov/sab> prior to the meeting.

Procedures for Providing Public Input: Interested members of the public may submit relevant written or oral information for consideration on the topics included in this advisory activity.

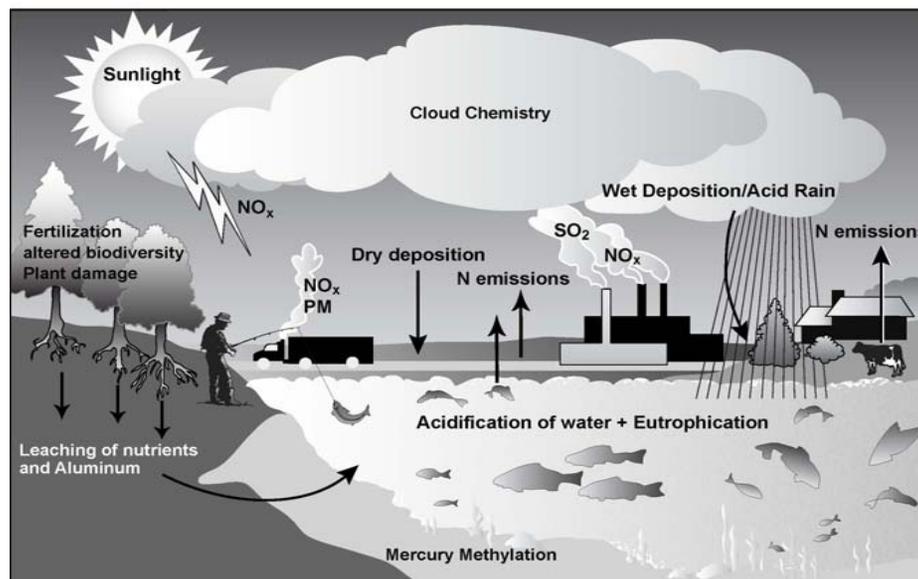
Oral Statements: To be placed on the public speaker list for the April 2-3, 2008 meeting, interested parties should notify Ms. Kyndall Barry, DFO, by e-mail no later than March 28, 2008. Oral presentations will be limited to one-half hour for all speakers. To be placed on the public speaker list for the May 5, 2008 teleconference, interested parties should notify Mr. Fred Butterfield, DFO, by e-mail no later than May 1, 2008. Oral presentations will be limited to a total of 30 minutes for all speakers.

Written Statements: Written statements for the April 2-3, 2008 meeting should be received in the SAB Staff Office by March 28, 2008 so that the information may be made available to the CASAC Panel for its consideration prior to this meeting. For the teleconference meeting of the chartered CASAC on May 5, 2008, statements should be received in the SAB Staff Office by May 1, 2008. Written statements should be supplied to the appropriate DFO in the following formats: one hard copy with original signature and one electronic copy via e-mail (acceptable file formats: Adobe Acrobat PDF, MS Word, WordPerfect, MS PowerPoint, or Rich Text files in IBM-PC/Windows 98/2000/XP format).

Accessibility: For information on access or services for individuals with disabilities, please contact Ms. Barry at the phone number or e-mail address noted above, preferably at least ten days prior to the face-to-face meeting, to give EPA as much time as possible to process your request.

Dated: February 15, 2008.  
Anthony F. Maciorowski,  
Deputy Director, EPA Science Advisory Board Staff Office.  
[FR Doc. E8-3613 Filed 2-25-08; 8:45 am]

# Background and History of the Review of the Secondary $\text{NO}_2$ and $\text{SO}_2$ NAAQS



Presentation to CASAC  
April 2, 2008

National Center for Environmental Assessment, Office of Research and Development  
Office of Air Quality Planning and Standards, Office of Air and Radiation

# Schedule

- Planning:
  - Workshop on science policy issues – July 2007
  - CASAC consultation on draft Integrated Review Plan – Oct. 2007
  - Final Integrated Review Plan – Dec. 2007
- April 2-3, 2008 CASAC meeting:
  - Review of 1<sup>st</sup> draft Integrated Science Assessment (ISA) and
  - Consult on draft Scope and Methods Plan for Risk/Exposure Assessment (R/EA)
- Next steps:
  - 2<sup>nd</sup> draft ISA and 1<sup>st</sup> draft R/EA release in Aug. 2008 -- CASAC review Oct. 2008
  - Final ISA – Dec. 12, 2008
  - 2<sup>nd</sup> draft R/EA release in Mar. 2009 – CASAC review May 2009
  - Final R/EA – July 2009
  - Rulemaking:
    - ANPR in Aug. 2009 -- CASAC review Oct. 2009
    - Proposed rule – Feb. 12, 2010
    - Final rule – Oct. 19, 2010

# Regulatory History of the Secondary NO<sub>2</sub> and SO<sub>2</sub> NAAQS

- NO<sub>2</sub> and SO<sub>2</sub> secondary standards set in 1971; only change has been revocation of annual secondary (but not primary) SO<sub>2</sub> standard. Current secondary standards include:
  - 0.053 ppm NO<sub>2</sub> annual arithmetic average, mean of 1-hr concentrations (same as primary); to protect against direct gas-phase effects to vegetation
  - 0.50 ppm SO<sub>2</sub>, 3-hr average (different from primary); to protect against direct short-term effects on trees (foliar injury)
- Previous reviews have discussed the importance of broader environmental effects of NO<sub>x</sub> and SO<sub>x</sub>, but EPA decided not to revise the secondary NAAQS on the basis of such effects
- Numerous assessments, reviews, statutory/regulatory activities, and other initiatives have been undertaken over the last 3 decades, attesting to the complexity of the relevant science and policy issues (see Integrated Plan discussion)

## In this review . . .

- Separate review of secondary standards from reviews of primary standards
- Take multi-pollutant approach, linking NO<sub>x</sub> and SO<sub>x</sub> as well as considering both oxidized and reduced forms of nitrogen
- Focus on environmental effects related to deposition of sulfur and reactive nitrogen into sensitive terrestrial and aquatic ecosystems
- Avoid unnecessary overlap across different NAAQS reviews
- Take a broad view of potential policy outcomes, after first evaluating relevant science and designing/conducting relevant assessments

## **Attachment E**

# **Integrated Science Assessment for Oxides of Nitrogen and Sulfur — Environmental Criteria (1st External Review Draft)**

**Presentation to the  
U.S. EPA Clean Air Scientific Advisory Committee**

***Nitrogen and Sulfur Oxides, Environmental Criteria Team***  
**National Center for Environmental Assessment, RTP Division**  
**Office of Research and Development**  
**U.S. EPA**

**2 April 2008**

# ***NCEA-RTP Nitrogen and Sulfur Oxides Team ISA in Support of the Secondary Standard***

**Dr. Ila Cote – Acting Division Director**

**Dr. Mary Ross – Branch Chief**

**Dr. Tara Greaver – NO<sub>x</sub> and SO<sub>x</sub> Environmental Criteria Team Leader**

**Dr. Jeffrey R. Arnold**

**Dr. Jean-Jacques B. Dubois**

**Dr. Jeffrey Herrick**

**Dr. Lingli Liu**

**Dr. Kristopher Novak**

**Dr. Paul F. Wagner**



## Presentation Overview

### Highlights from the draft ISA organized by CASAC charge questions

- Descriptions of sources, transformations, and ecological exposures
  - **NO<sub>x</sub>, SO<sub>x</sub>, and NH<sub>x</sub>** atmospheric chemistry and physics
  - Characterization of ambient concentrations
  - Characterization of deposition totals and methods for computing them
  
- Characterizations of ecological effects
  - **Acidification**
    - Occurs in response to atmospheric deposition of NO<sub>x</sub> and SO<sub>x</sub>
  - **Nitrogen enrichment**
    - Occurs in response to atmospheric deposition NO<sub>x</sub>, NH<sub>x</sub> and other forms of reactive nitrogen (Nr)
  - Other welfare effects
    - Stimulation of Hg methylation by atmospheric deposition of SO<sub>4</sub><sup>2-</sup>
    - Direct phytotoxic effects of gas-phase NO<sub>x</sub> and SO<sub>x</sub>



## **Charge Questions 1-3: Atmospheric Science and Exposure**

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  $\text{NO}_x$  and  $\text{SO}_x$ ?
2. How well characterized are the relevant properties of the ambient air concentrations and deposition of  $\text{NO}_x$  and  $\text{SO}_x$ , including policy-relevant background (PRB) 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?



## ***NO<sub>x</sub> and NH<sub>x</sub> Emissions***

**NO<sub>x</sub> emissions (chiefly NO+NO<sub>2</sub>)** are split roughly evenly between all point and area stationary sources together, and all mobile sources

- 2002 total U.S. emissions  $\approx$  23.2 Tg

**Biogenic additions of NO<sub>x</sub>** from biomass burning, soil off-gassing, and lightning are **substantially smaller fractions** of the budget

- NO and N<sub>2</sub>O from soils as intermediate products of denitrification either naturally or after N fertilizers added
- NO<sub>x</sub> from managed agriculture and forests  $\approx$  0.01 Tg in 2002
- N<sub>2</sub>O was  $\sim$ 6.5% of total U.S. greenhouse gas emissions on a Tg CO<sub>2</sub> equivalent basis in 2005, with >75% emitted from agricultural soils

**NH<sub>3</sub> emissions chiefly from livestock and soils** after N fertilization

- 2002 NH<sub>3</sub> emissions from all U.S. sources  $\approx$  4 Tg, with >85% from agricultural and silvicultural sectors



## *Ambient and Background NO<sub>x</sub>*

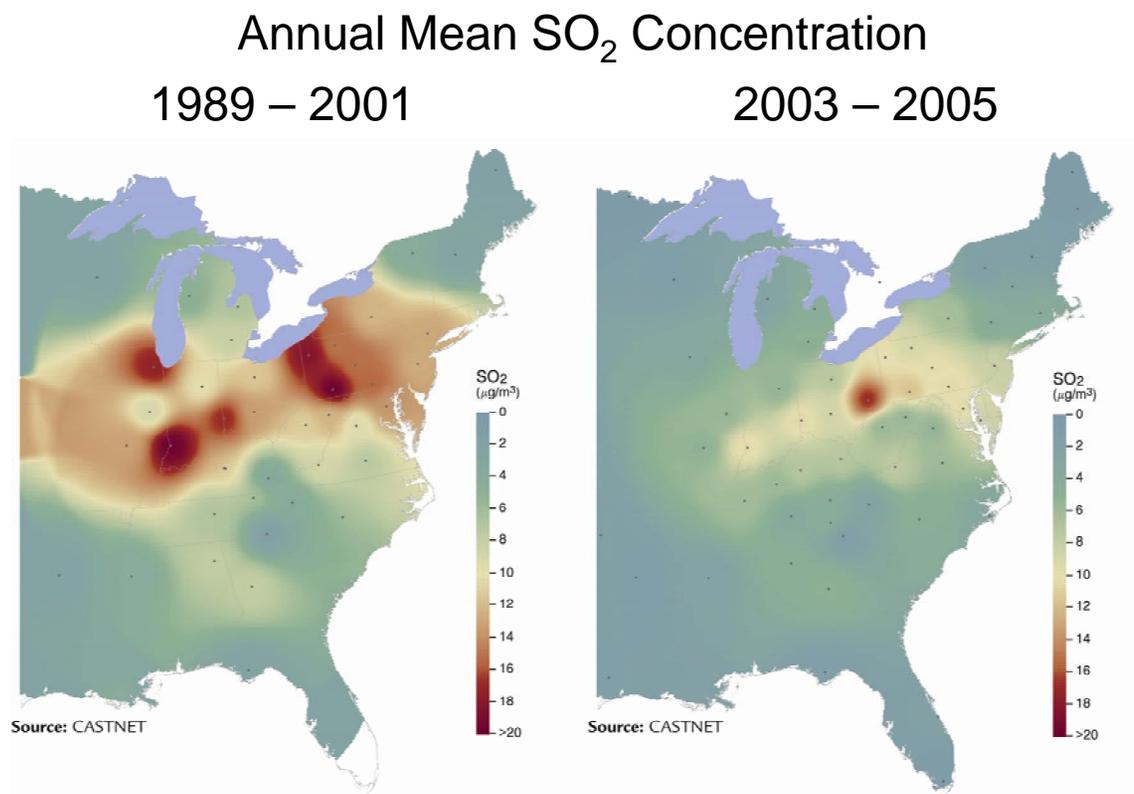
In the U.S. for the years 2003–2005

- **24 h average ambient NO<sub>2</sub> mixing ratios** in cities, where most NO<sub>x</sub> is produced
  - **<20 parts per billion (ppb)** with a 99<sup>th</sup> percentile value <50 ppb
- **Annual-average NO<sub>2</sub> mixing ratios** at nearly all urban, rural, and remote monitored sites
  - **<5 ppb**
- Annual-average **policy-relevant background NO<sub>2</sub>** mixing ratios are computed to be:
  - **<300 parts per trillion (ppt)** over most of the continental U.S.



## Ambient and Background SO<sub>2</sub>

The national composite annual mean ambient SO<sub>2</sub> concentrations have decreased by 48% from 1990 to 2005

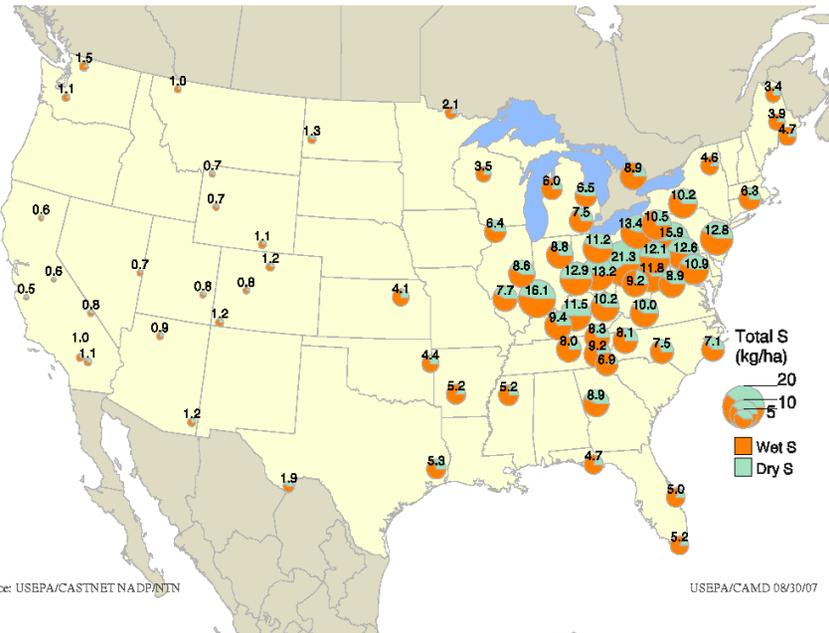


2005 mean ambient SO<sub>2</sub> mixing ratio: ~4 ppb  
Background (excepting Pacific Northwest): ~10 to 30 ppt

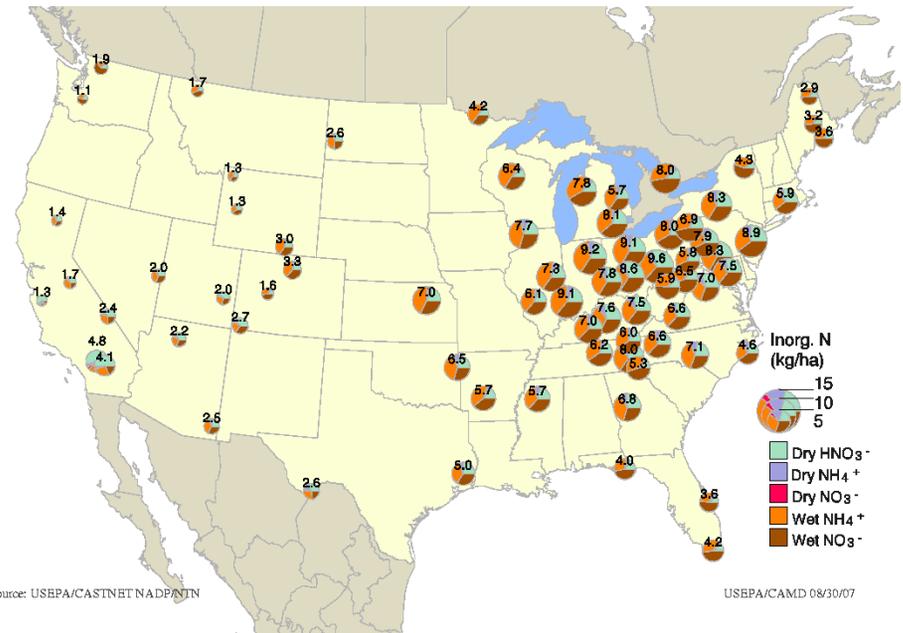


# Annual-average Deposition, 2004–2006

## Total S



## Inorganic N



Thin coverage of monitoring sites leaves us blind in many areas, especially in the west

Some special study measurements and numerical modeling experiments suggest that hotspots can be missed by routine monitors

- Measured N deposition is  $>20 \text{ kg ha}^{-1} \text{ y}^{-1}$  in some regions of the NY Adirondacks
- Model estimates as high as  $32 \text{ kg ha}^{-1} \text{ y}^{-1}$  for a region of southern CA with more than half predicted to come from  $\text{NO}_x$



## **Charge Questions 4-6: Ecosystem Acidification**

4. How well are the major effects of  $\text{NO}_x$  and  $\text{SO}_x$  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  $\text{NO}_x$  and  $\text{SO}_x$  and environmental effects?
6. How well characterized is the relative importance of the oxidized and the reduced forms of nitrogen on ecosystem acidification?



## *Acidification*

**SO<sub>x</sub> deposition** is the main cause of chronic surface water acidification

- The good news: 1/4 to 1/3 of lakes and streams chronically acidic in the 1980s were no longer chronically acidic in the year 2000; largely attributed to decreases in sulfur deposition
- The bad news: Accumulation in soil due to historic loading in addition to current loading inhibits the recovery of some regions

**NO<sub>x</sub> deposition** is an important cause of episodic acidification

- The Episodic Research Program demonstrated that episodic acidification has long-term adverse effects on biota, especially fish populations



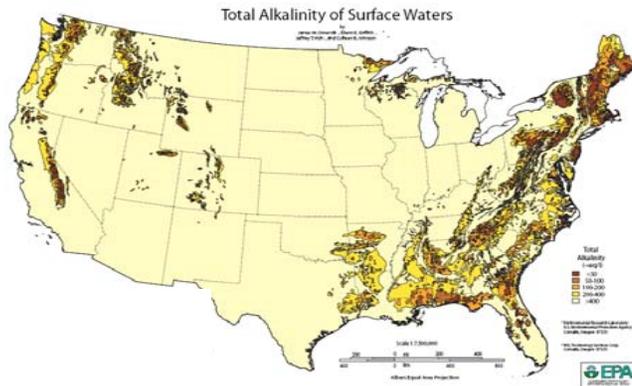
## Acidification

Toxic effects on terrestrial ecosystems include:  
Al<sup>3+</sup> toxicity and lower cold tolerance that lead to decreased growth and mortality of tree species

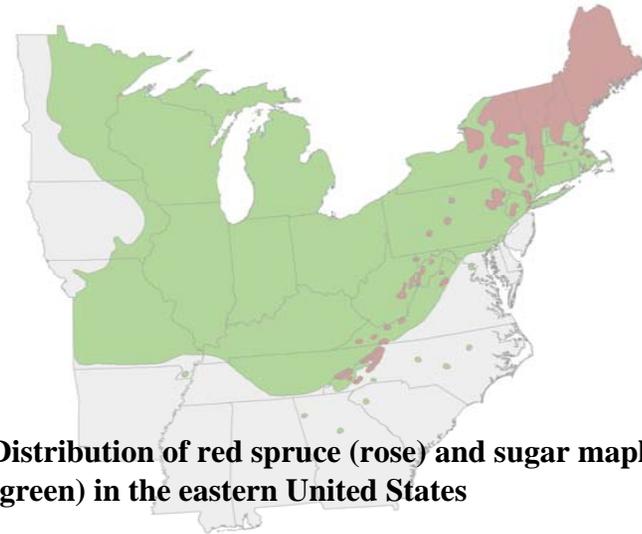
- Red spruce and sugar maple, especially at high elevation

Toxic effects on aquatic ecosystems include:

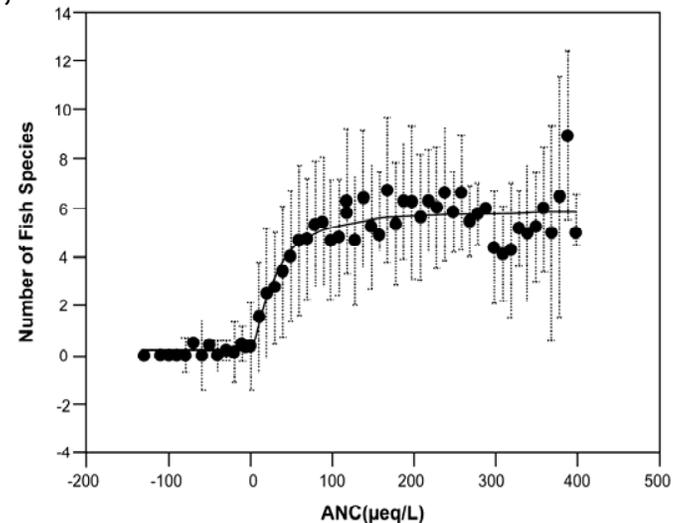
Mortality across trophic levels including phytoplankton, zooplankton, macroinvertebrates, and fish; few studies on higher trophic levels



Darker colors indicate greater acid sensitivity



Distribution of red spruce (rose) and sugar maple (green) in the eastern United States



Number of fish species per lake versus acidity status, expressed as ANC, for Adirondack lakes



# Chemical Indicators of Acidic Deposition

## CHEMICAL INDICATORS OF EFFECTS ON AQUATIC ECOSYSTEMS

| Chemical Indicator  | Potential Threshold   | References                                                                   |
|---------------------|-----------------------|------------------------------------------------------------------------------|
| • Surface water pH  | 5.0-6.0               | Baker et al., 1990                                                           |
| • Surface water ANC | 0-50 $\mu\text{eq/L}$ | Bulger et al., 1999                                                          |
| • Inorganic Al      | 2-4 $\mu\text{mol/L}$ | Wigington Jr. et al., 1996<br>Driscoll et al., 2001;<br>Baldigo et al., 2007 |

## CHEMICAL INDICATORS OF EFFECTS ON TERRESTRIAL ECOSYSTEMS

| Chemical Indicator          | Potential Threshold | References                                                              |
|-----------------------------|---------------------|-------------------------------------------------------------------------|
| • Soil base saturation      | 10-20%              | Lawrence et al., 2006;<br>Driscoll et al., 2001;<br>Cronan et al., 1990 |
| • Soil solution Ca:Al ratio | 1.0                 | Cronan and Grigal, 1995                                                 |
| • Soil C:N ratio            | 20-25               | Aber et al., 2003                                                       |



# Biological Indicators of Acidic Deposition

## BIOLOGICAL INDICATORS OF EFFECTS ON AQUATIC ECOSYSTEMS

| Indicator                                                                                     | Measures                                                                      | References                                    |
|-----------------------------------------------------------------------------------------------|-------------------------------------------------------------------------------|-----------------------------------------------|
| <ul style="list-style-type: none"><li>Fishes, zooplankton<br/>crustaceans, rotifers</li></ul> | Condition factor<br>Presence/absence<br>Diversity metrics<br>Tolerance values | Baker et al., 1990b<br>Sullivan et al., 2006b |

## BIOLOGICAL INDICATORS OF EFFECTS ON TERRESTRIAL ECOSYSTEMS

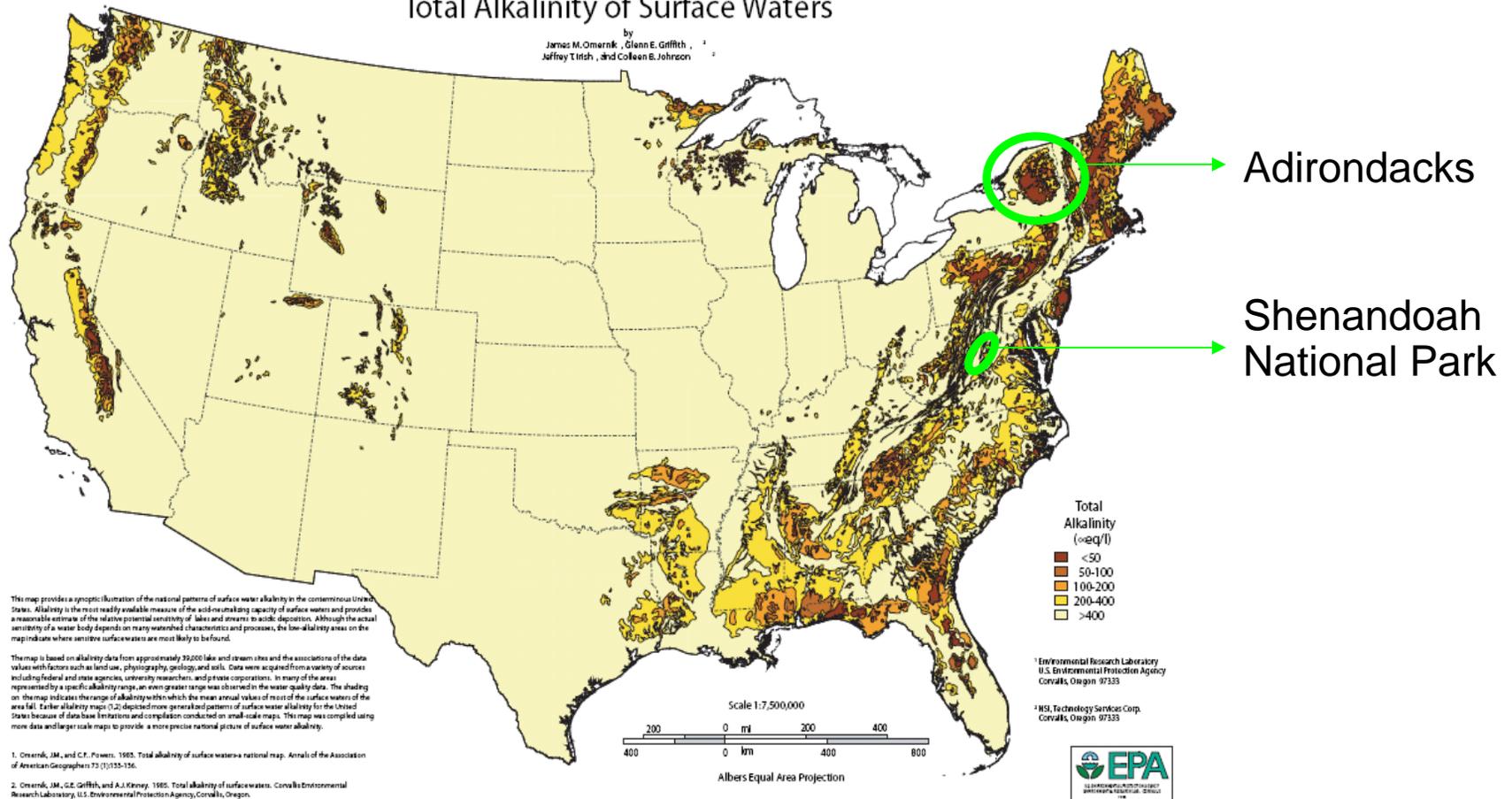
| Indicator Species                                           | Example of Health Indices                                                    | References                                       |
|-------------------------------------------------------------|------------------------------------------------------------------------------|--------------------------------------------------|
| <ul style="list-style-type: none"><li>Red spruce</li></ul>  | Percent dieback of canopy trees                                              | Shortle et al., 1997;<br>DeHayes et al., 1999    |
| <ul style="list-style-type: none"><li>Sugar maple</li></ul> | Basal area dead sugar maple (as %)<br>Crown vigor index<br>Fine twig dieback | Bailey et al., 1999;<br>Drohan and DeWalle, 2002 |



# Regional Sensitivity to Acidification and Selected Impaired Ecosystems under Current Deposition Levels

## Total Alkalinity of Surface Waters

by  
James M. Omernik, Glenn E. Griffith,  
Jeffrey T. Irish, and Colleen B. Johnson



This map provides a synoptic illustration of the national patterns of surface water alkalinity in the conterminous United States. Alkalinity is the most readily available measure of the acid-neutralizing capacity of surface waters and provides a reasonable estimate of the relative potential sensitivity of lakes and streams to acidic deposition. Although the actual sensitivity of a water body depends on many watershed characteristics and processes, the low-alkalinity areas on the map indicate where sensitive surface waters are most likely to be found.

The map is based on alkalinity data from approximately 30,000 lake and stream sites and the association of the data values with factors such as land use, physiography, geology, and soils. Data were acquired from a variety of sources including federal and state agencies, university researchers, and private corporations. In many of the areas represented by a specific alkalinity range, an even greater range was observed in the water quality data. The shading on the map indicates the range of alkalinity within which the mean annual values of most of the surface waters of the area fall. Earlier alkalinity maps (1,2) depicted more general and patterns of surface water alkalinity for the United States because of data base limitations and compilation conducted on small-scale maps. This map was compiled using more data and larger scale maps to provide a more precise national picture of surface water alkalinity.

1. Omernik, J.M., and C.F. Powers. 1983. Total alkalinity of surface waters: national map. *Annals of the Association of American Geographers* 73 (1):135-156.

2. Omernik, J.M., G.E. Griffith, and A.J. Minery. 1985. Total alkalinity of surface waters. Corvallis Environmental Research Laboratory, U.S. Environmental Protection Agency, Corvallis, Oregon.



RESEARCH & DEVELOPMENT

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## Case Studies

### Adirondacks

- Overall improvements in lakewater acid-base chemistry have been modest
- Modeling results suggested that recovery for the most acid-sensitive Adirondack lakes would not continue

### Shenandoah

- Modeling results for the Southern Appalachian Mountains region, south of Virginia and West Virginia, suggested that, under current emissions levels, the percentages of acidic streams ( $\text{ANC} < 0 \mu\text{eq l}^{-1}$ ) will increase
- Simulations suggested that re-acidification might be prevented with deposition (Sullivan et al., 2007a)



## **Charge Questions 7-9: Ecosystem Nitrogen Enrichment**

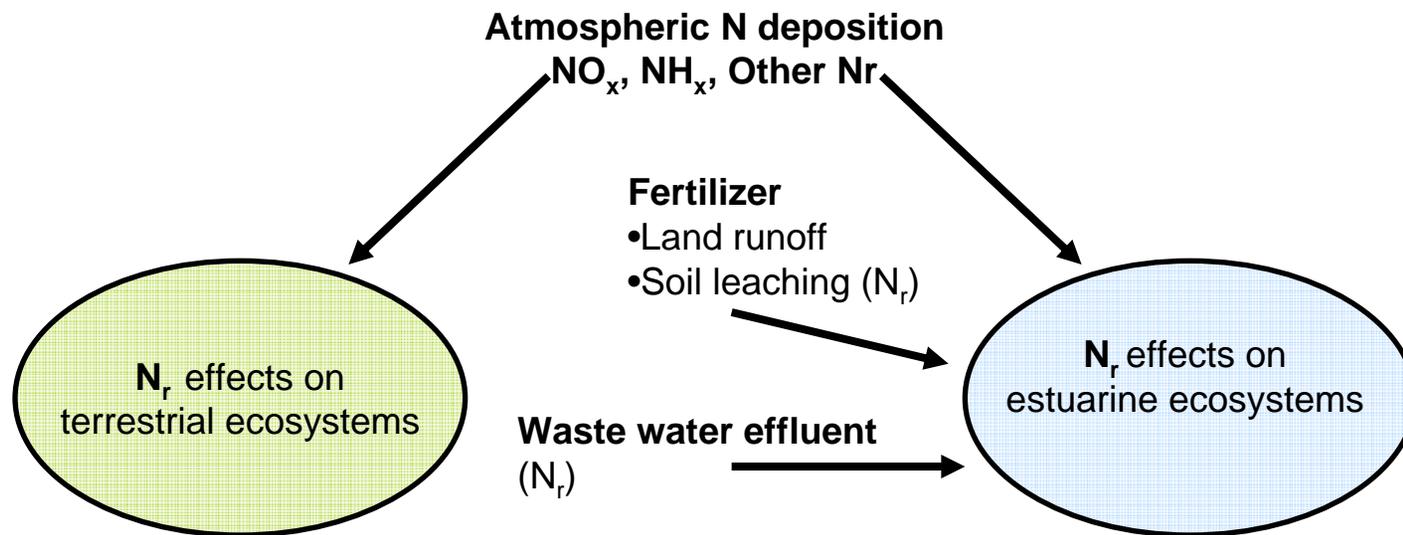
7. How well are the major effects of NO<sub>x</sub> as it contributes to nitrogen enrichment of the ecosystems appropriately identified and characterized? To what extent do the discussions and integration of evidence across scales (e.g., various species, communities, ecosystems, and regions) correctly 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?



# Nitrogen Enrichment

Atmospheric N deposition causes a cascade of ecological effects at multiple scales

- At the smallest scale is the increased growth of individual species
- Not all species can take advantage of the additional N; some lose their competitive advantage
- N additions cause a suite of terrestrial and aquatic ecological problems including biodiversity losses, community shifts, eutrophication, and harmful algal blooms



# Terrestrial & Freshwater Ecosystem Indicators

## Chemical indicators

- Soil C:N ratio
- Soil water [NO<sub>3</sub><sup>-</sup>]
- Nitrate leaching
- Chlorophyll a
- Chlorophyll a: Total P
- Water [NO<sub>3</sub><sup>-</sup>]
- Dissolved inorganic N (DIN)

## Biological Indicators

- Altered community composition, biodiversity and /or population decline
  - Diatom species
  - Lichen species
  - Mycorrhizal species
  - Moss species
  - Grass and herbaceous species
  - Butterfly species
- Foliar/plant tissue [N], C:N, N:Mg, N:P
- Phytoplankton biomass/production
- Terrestrial plant biomass/production



## Nitrogen Nutrient Enrichment: Terrestrial, Wetland and Freshwater Aquatic

(kg N ha<sup>-1</sup> yr<sup>-1</sup>)

~1.5

Altered **diatom** communities in high elevation freshwater lakes  
Elevated **N in tree leaf tissue** high elevation forests (Colorado; Baron, 2000; Baron, 2006; Saros et al., 2003)

3 to 8

Mortality of sensitive **lichen** species (Pacific NW; Geiser and Neitlich, 2007)

5 to 35

**Species richness** declines as a linear function of the rate of inorganic nitrogen deposition, with a reduction of one species per 4 m<sup>2</sup> quadrant for every 2.5 kg N yr<sup>-1</sup> deposition (U.K.; Stevens et al., 2004)

<6.3 to 10

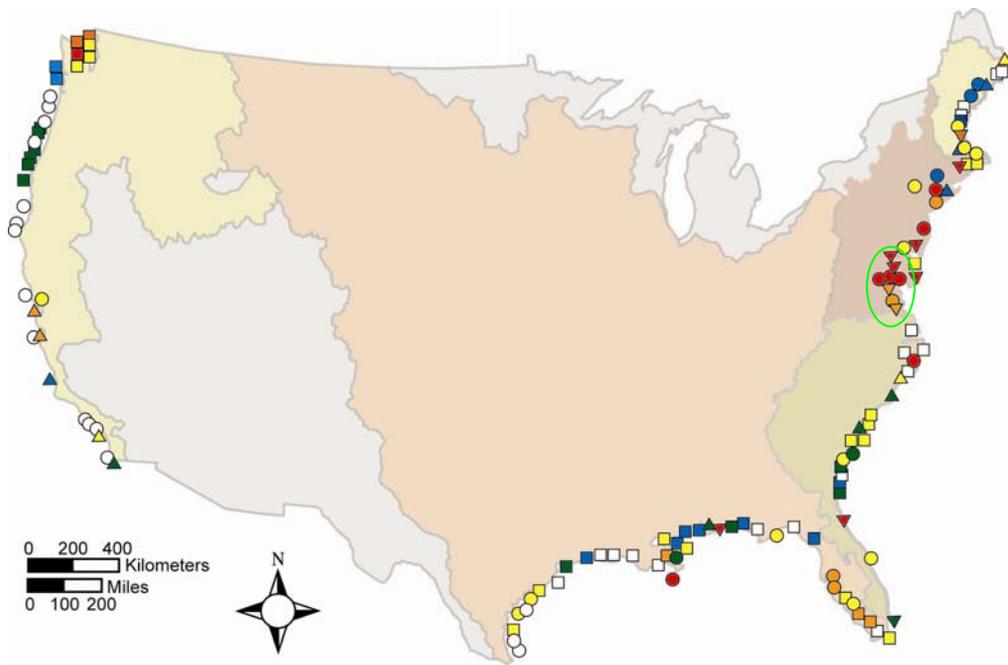
Onset of **nitrate leaching in many U.S. forests** (Aber et al., 2003)

<10 to 15

**Altered community composition** in native grasslands contributing to decline of native butterfly populations (California; Fenn et al., 2003; Weiss, 1999)



# Nitrogen Nutrient Enrichment: Estuary Eutrophication



- **High:** symptoms occur periodically or persistently and/or over an extensive area.
- **Moderate high:** symptoms occur less regularly and/or over a medium to extensive area.
- **Moderate:** symptoms occur less regularly and/or over a medium area.
- **Moderate low:** symptoms occur episodically and/or over a small to medium area.
- **Low:** few symptoms occur at more than minimal levels.
- Unknown:** insufficient data for analysis.

#### Change in eutrophic condition since 1999 assessment

- △ Symptoms **improved** since 1999 assessment.
- No change** in symptoms since 1999 assessment.
- ▽ Symptoms **worsened** since 1999 assessment.
- Insufficient data** to show trend

2007 *National Estuarine Eutrophication Assessment* evaluated 138 U.S. estuaries:

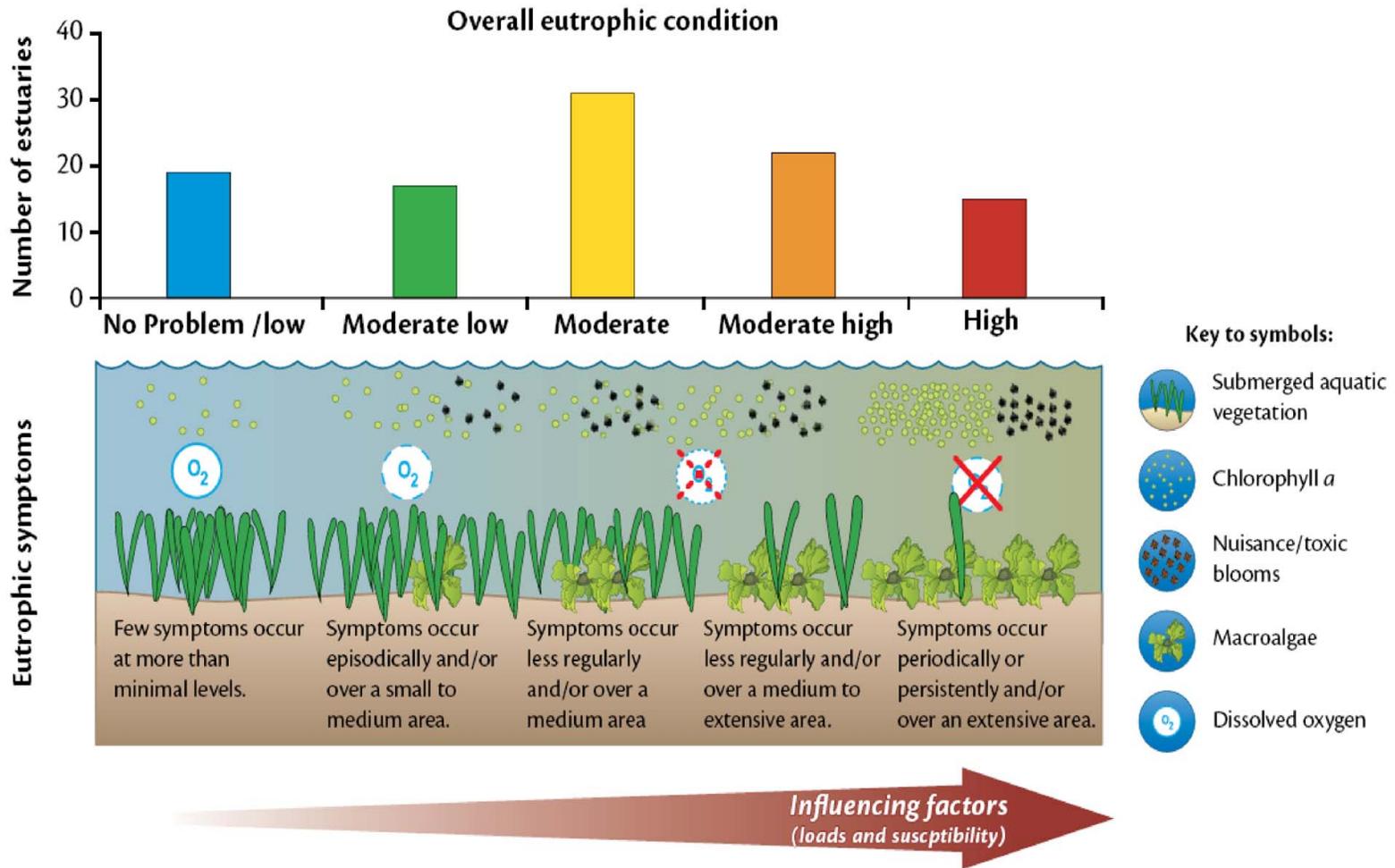
- 84 systems had moderate to highly eutrophic conditions

**Contribution of atmospheric N deposition to total N load has been modeled for some estuaries:**

- >30% for Chesapeake
- >70% for other estuaries



# Ecological Indicators of Eutrophication



## Charge Question 10

10. Several additional effects are discussed, including mercury methylation, direct gas-phase effects on foliage, and N<sub>2</sub>O as a greenhouse gas. How well does the draft ISA characterize the evidence on these topics?



## Other welfare effects

### Direct phytotoxic effects

- Gas-phase  $\text{SO}_2$  can cause acute foliar injury and decrease growth of plants. However, research on these effects has been limited in the past few decades
- Gas-phase  $\text{NO}_x$  has potential phytotoxic effects. The 1993  $\text{NO}_x$  AQCD concluded that concentrations of  $\text{NO}_2$  and  $\text{NO}$  in the atmosphere are rarely high enough for this. Very little new research has been done to alter this conclusion

Mercury, a neurological and reproductive toxin, enters food webs and bioaccumulates at higher trophic levels in the methylated form (MeHg). Sulfur reducing bacteria are the principal agent of Hg methylation, and  $\text{SO}_4^{2-}$  deposition increases their activity, and MeHg production

$\text{N}_2\text{O}$  is a greenhouse gas



## **Charge Questions 11-12**

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?



## Summary Points

The emissions and atmospheric concentrations of most N and S compounds are better characterized than their deposition fields across the landscape

- Networks for measuring deposition are insufficiently dense to characterize fully the regional heterogeneity and hotspots revealed in field and modeling experiments
- Important components of total N deposition like  $\text{NH}_3$  are missing

Adverse ecological effects are due to the N and S deposition from current atmospheric concentrations of  $\text{NO}_x$  and  $\text{SO}_x$ , depending on the biological response considered

- Together, N and S deposition at current levels causes acidification of ecosystems in many regions
  - $\text{Al}^+$  toxicity and lower cold tolerance that lead to decreased growth and mortality of tree species (e.g. red spruce and sugar maple)
  - Mortality across trophic levels including phytoplankton, zooplankton, macroinvertebrates, and fish; few studies on higher trophic levels



## Summary Points

- N deposition from  $\text{NO}_x$  and  $\text{NH}_x$  sources at current levels contributes to adversity caused by excess nutrient enrichment
  - Atmospheric deposition of N to estuarine ecosystems may be 10 to >40% of the total N loadings
  - Deposition levels to terrestrial and freshwater ecosystems that range from 2-10  $\text{kg N ha}^{-1} \text{ yr}^{-1}$  cause the onset of effects



Attachment F

# Overview of the Scope and Methods Plan Supporting the Review of the Secondary NO<sub>2</sub>/SO<sub>2</sub> NAAQS



Anne Rea, Ph.D.

Presentation to CASAC

April 3, 2008

Office of Air Quality Planning and Standards



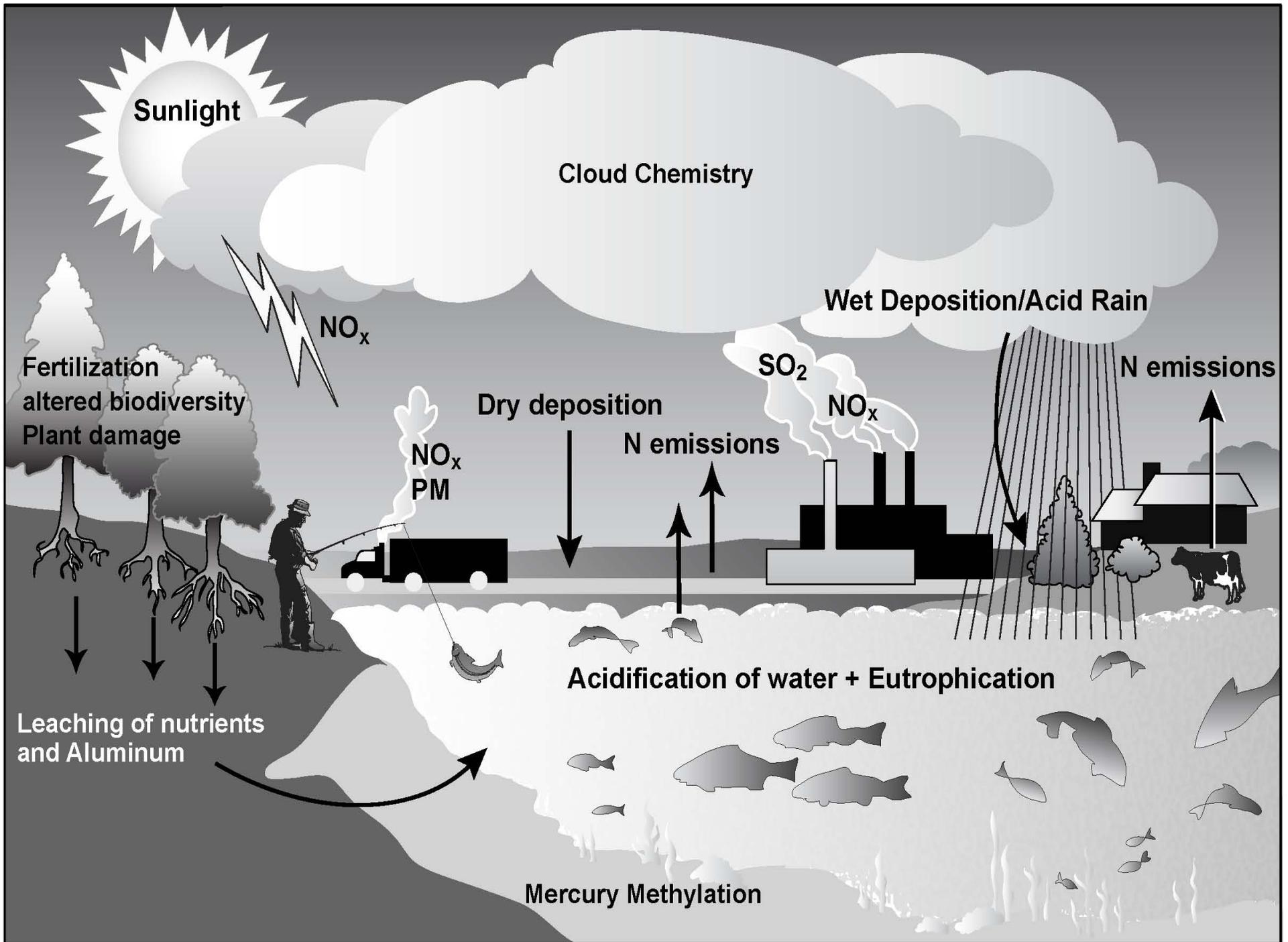
# Purpose

- Solicit feedback on EPA's planned approach to the NO<sub>x</sub>/SO<sub>x</sub> Secondary NAAQS Risk Assessment
  - Obtain guidance on breadth of the proposed risk assessment, which effects and case study areas should be targeted for this review

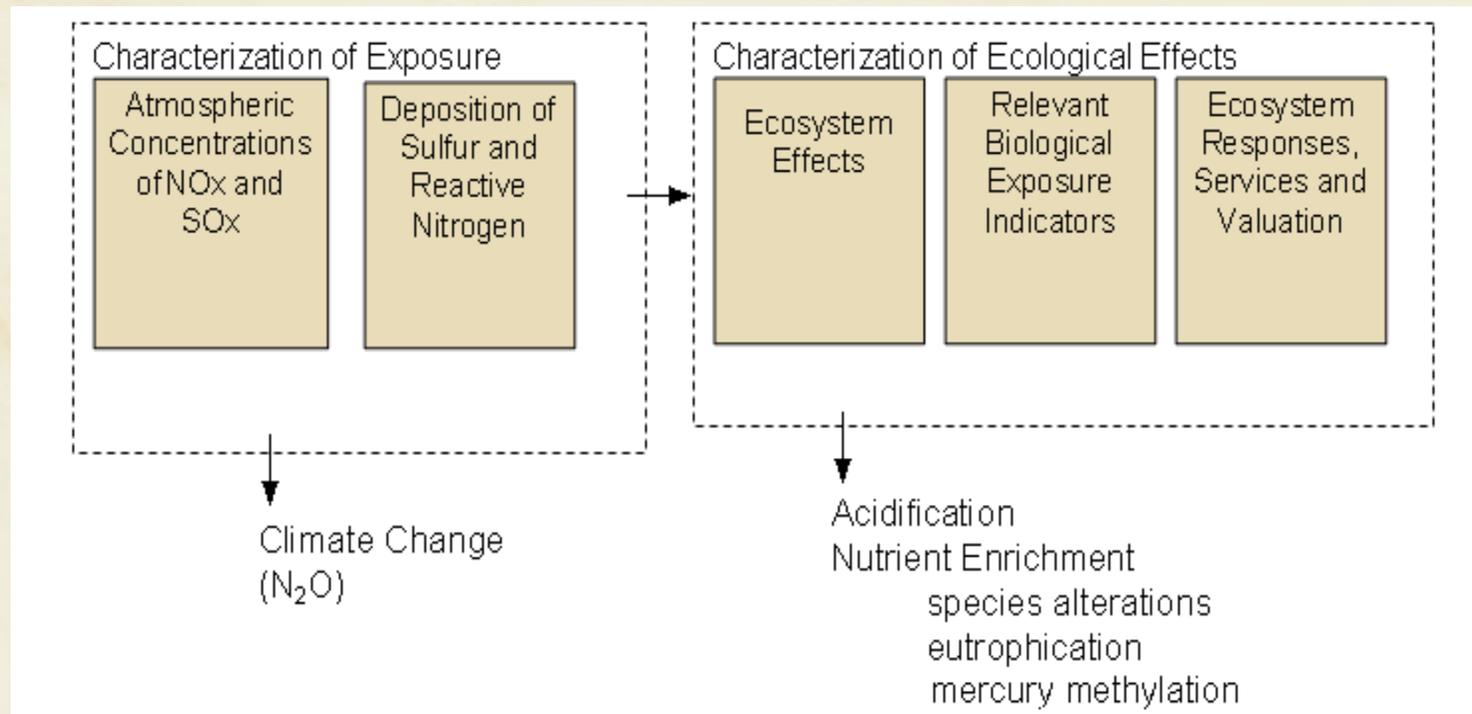


# Overview

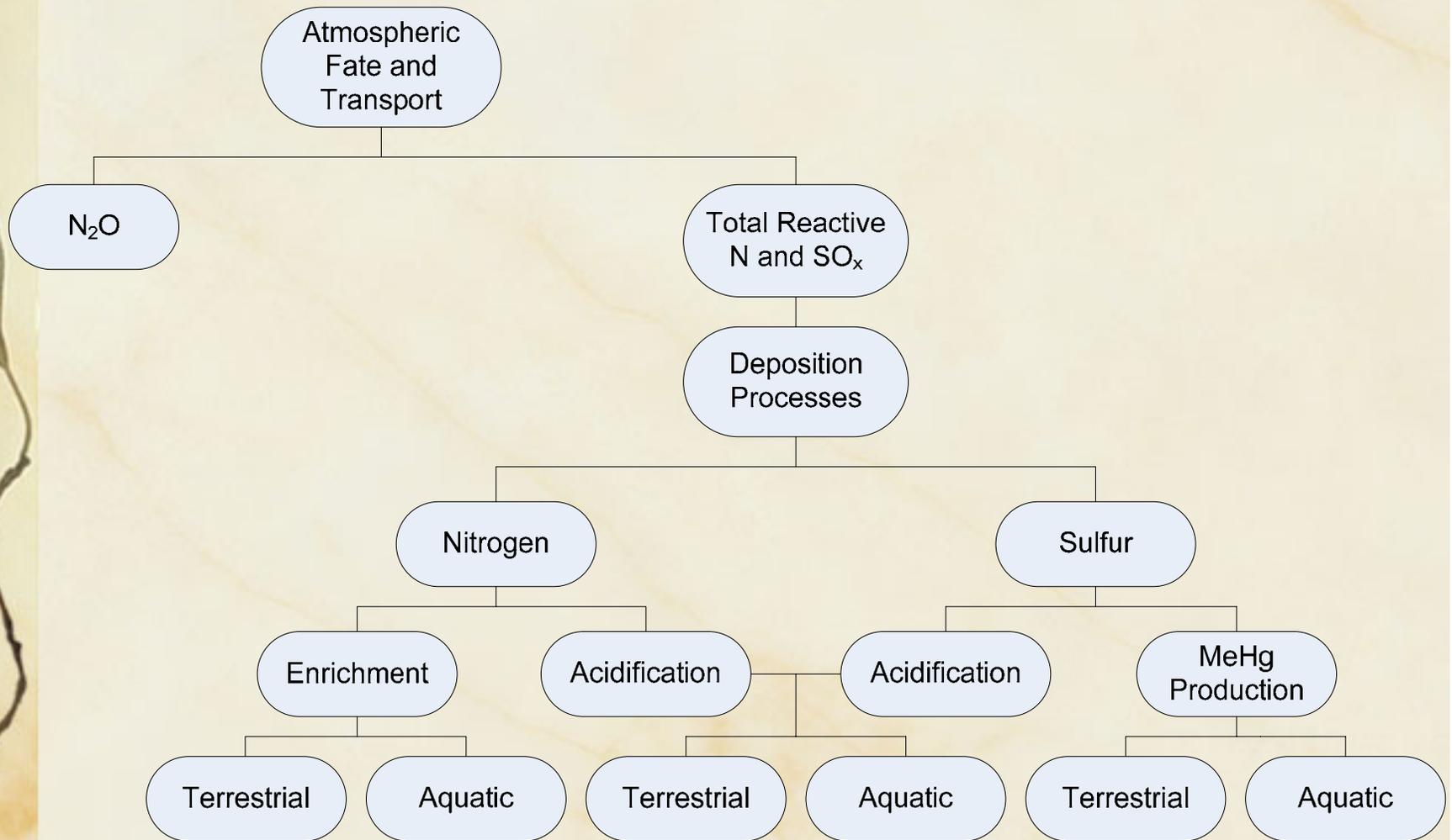
- Risk/Exposure Assessment
  - Overview of Risk Assessment Framework
  - Targeted Effects
    - Acidification
    - Nutrient Enrichment

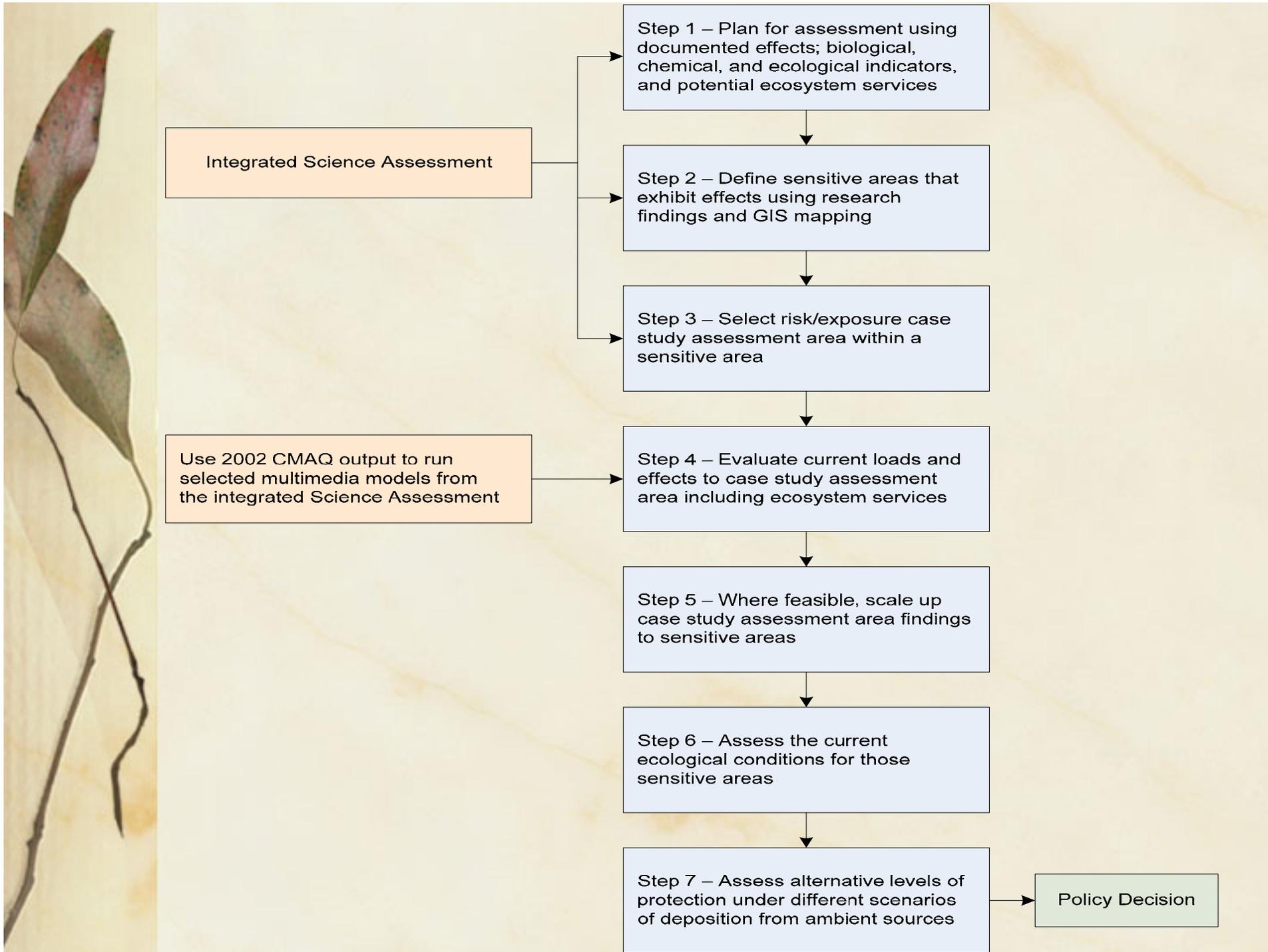


# Risk Assessment Framework



# Targeted Effects







# Where are we now?

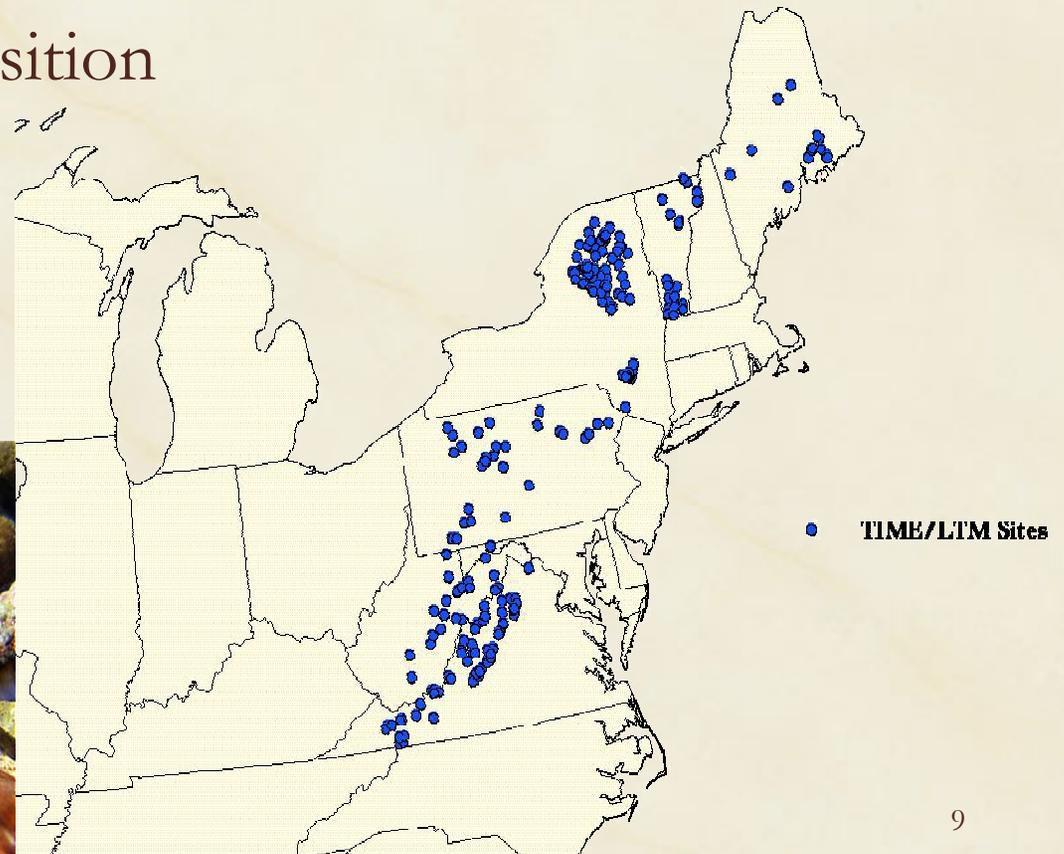
- Four main effect areas:
  - Aquatic acidification
  - Terrestrial acidification
  - Aquatic nutrient enrichment
  - Terrestrial nutrient enrichment
- Timeline
  - First draft RA to CASAC August, 2008
  - Second draft RA to CASAC March, 2009
  - Final RA July, 2009

# Aquatic Acidification

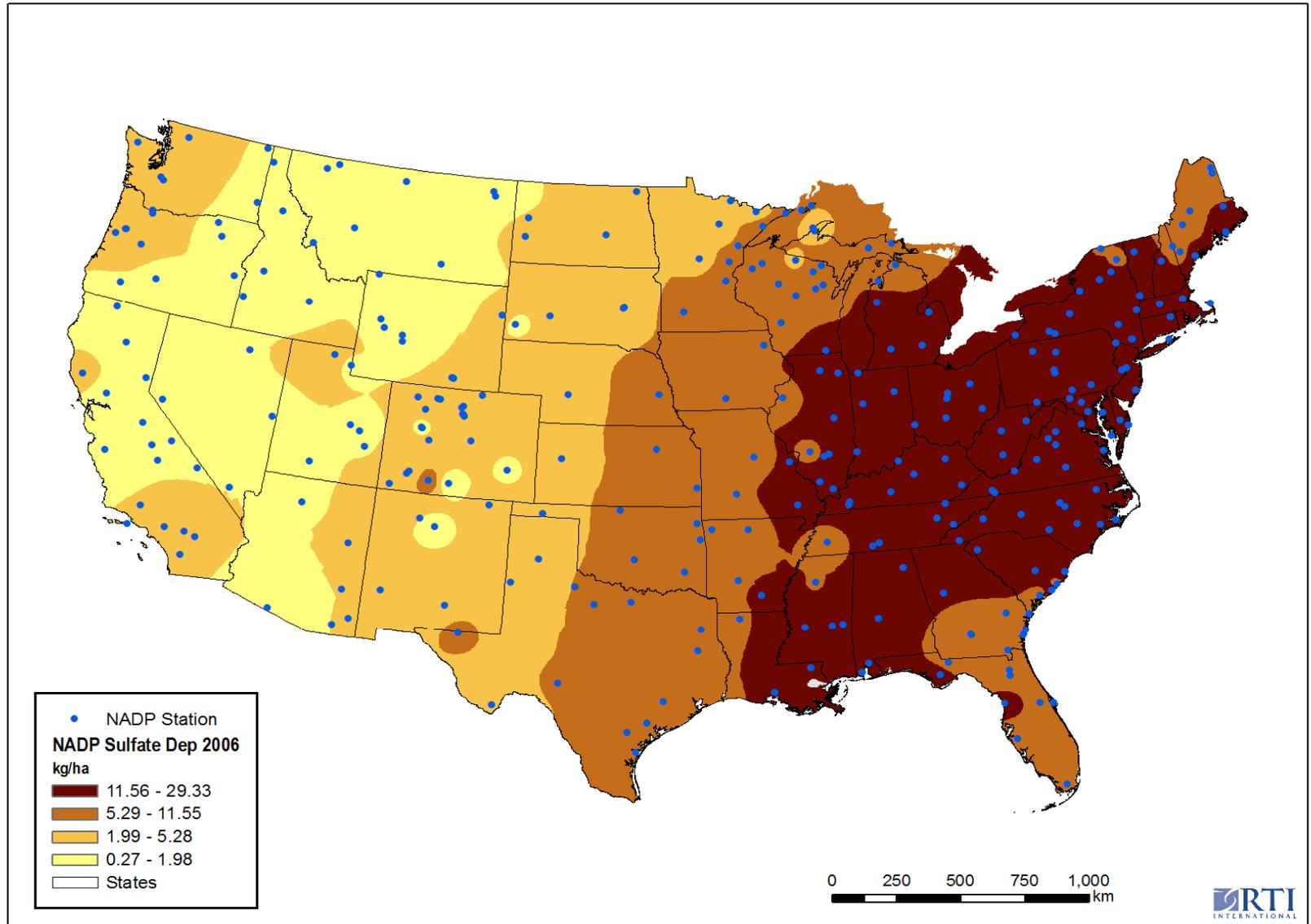
Examine ANC results and relationship to fish health

2002 CMAQ deposition

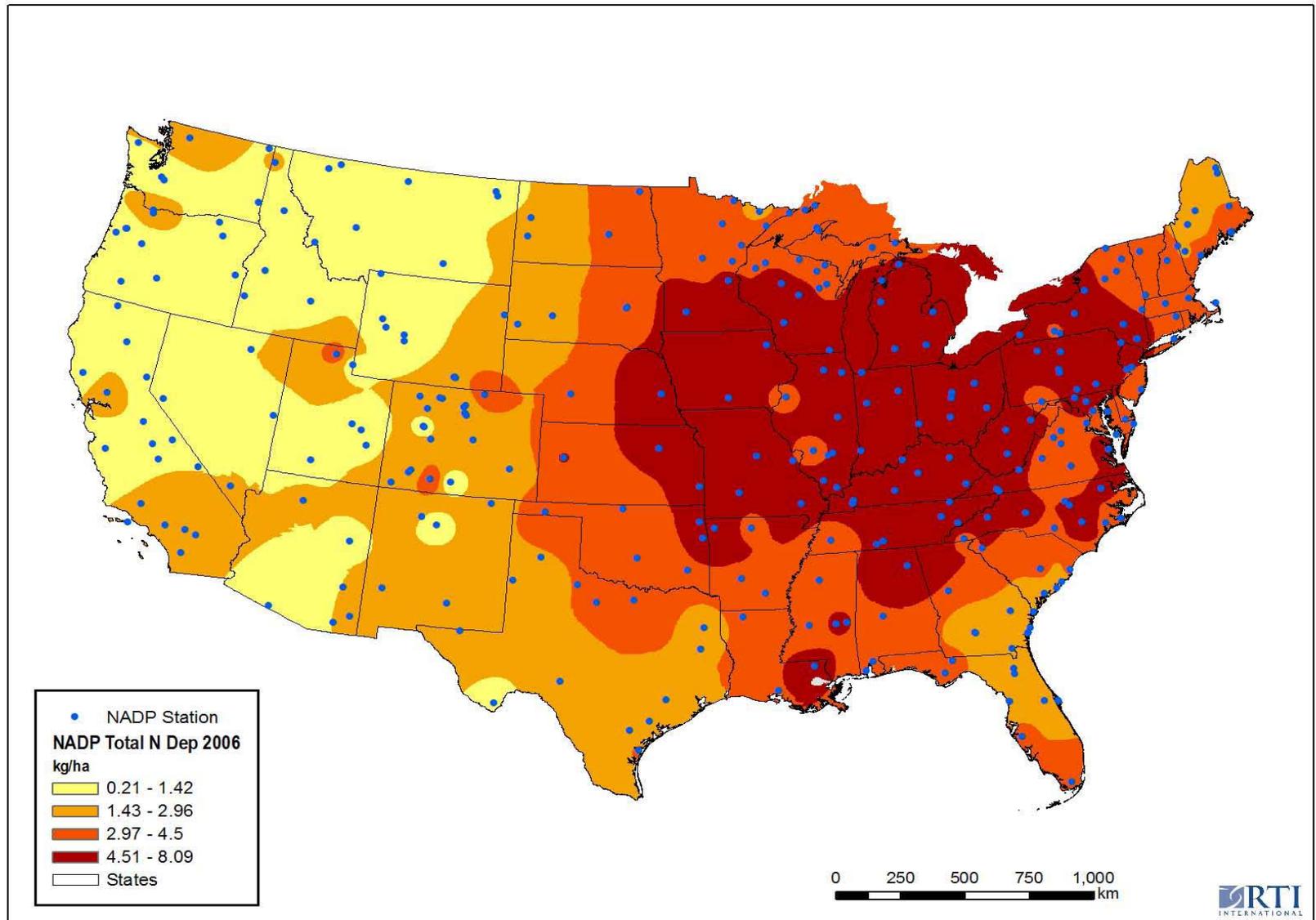
MAGIC model



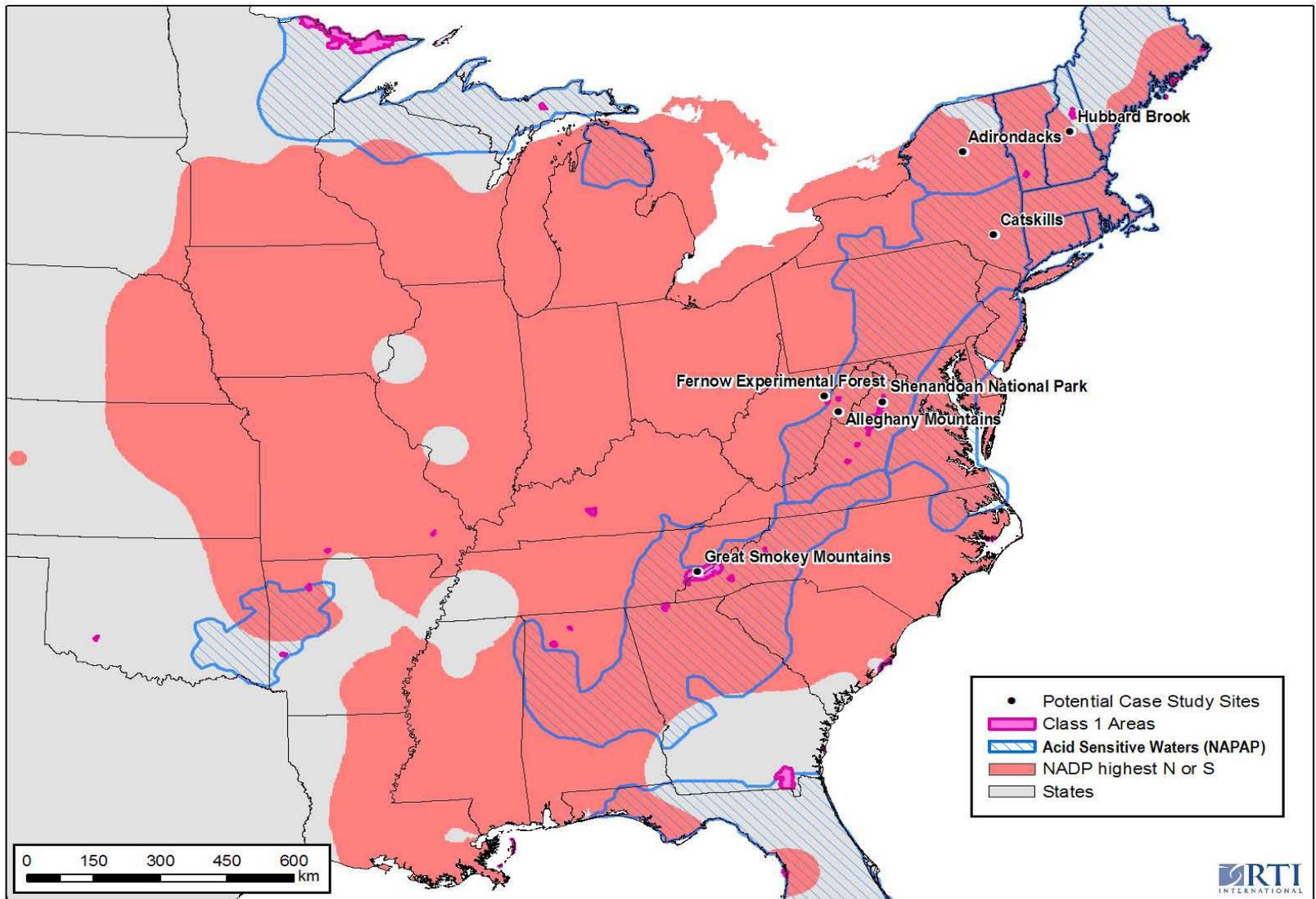
# Sulfur Deposition (NADP)



# Nitrogen Deposition (NADP)



# Aquatic Acid Sensitivity

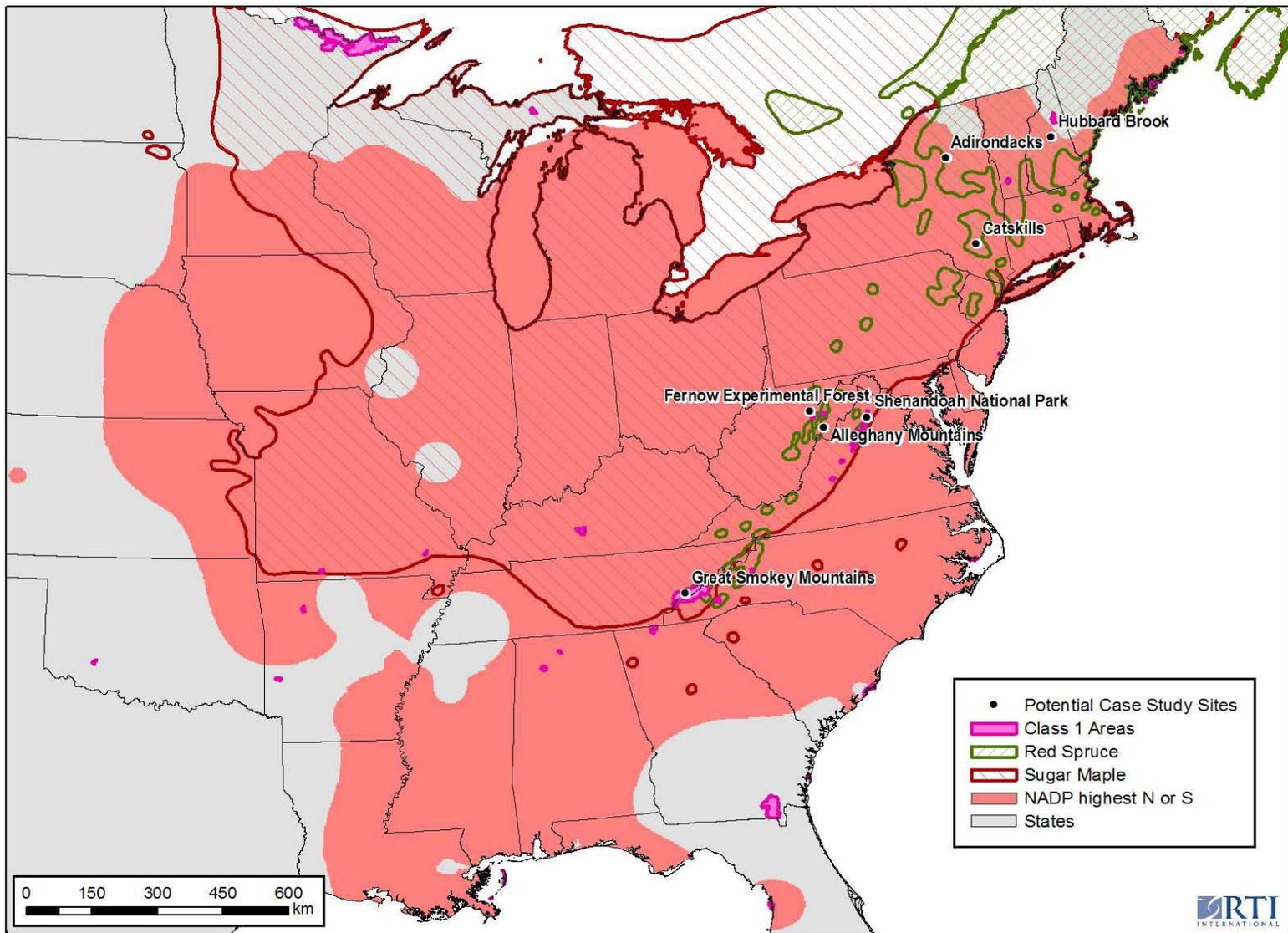


# Terrestrial Acidification

- Documented effects of acidification on red spruce and sugar maple
- Use the Simple Mass Balance model to examine changes in soil base chemistry
  - If possible, correlate to tree health



# Terrestrial Acid Sensitivity

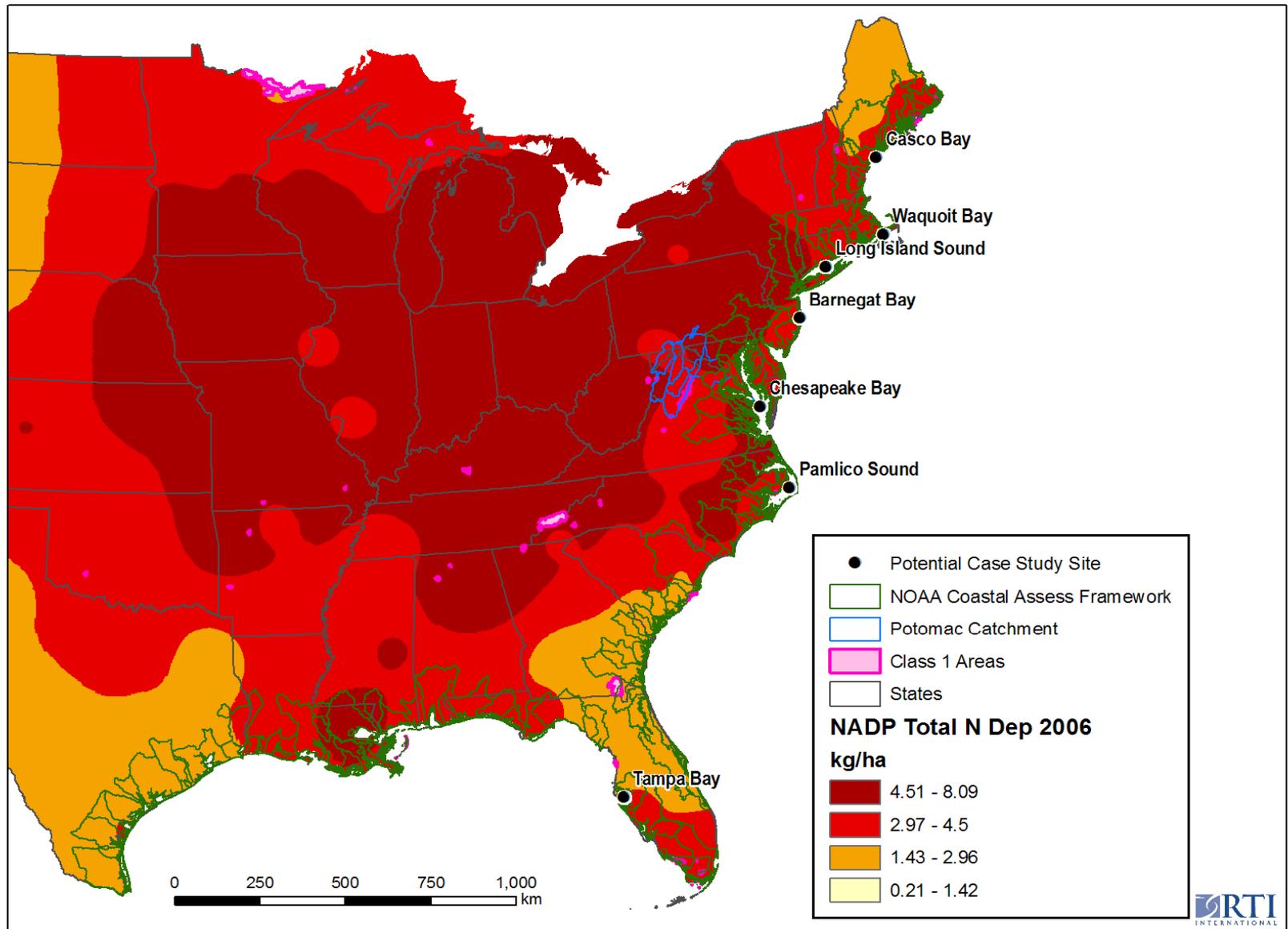


# Aquatic Nitrogen Enrichment

- East and Gulf Coast estuaries show significant effects, however are difficult to model
- NOAA eutrophication indices
- Model main stem river of one or more estuaries
- Also considering using DayCent-Chem model on some alpine lakes in the West



# Aquatic Nitrogen Enrichment

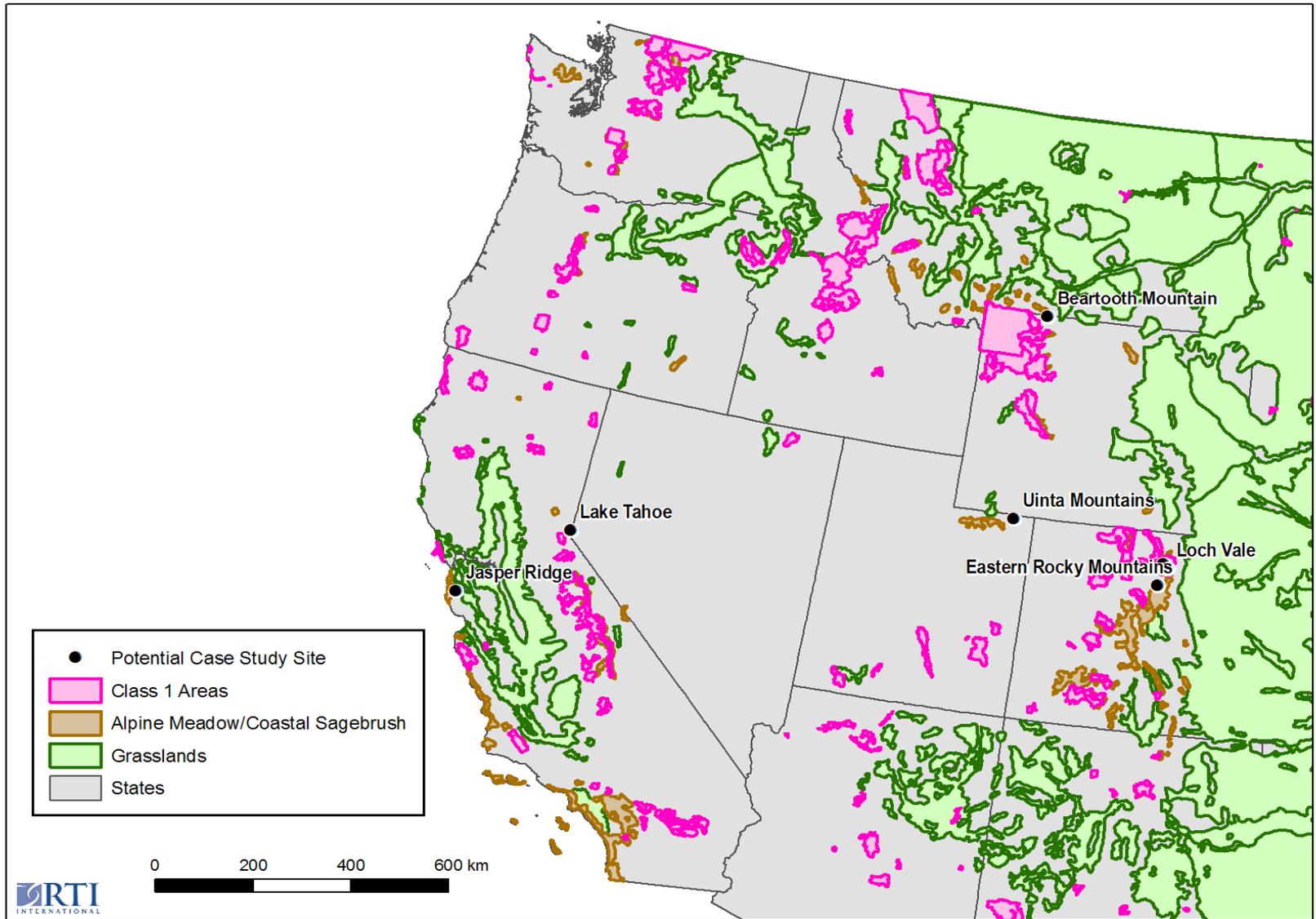


# Terrestrial Nitrogen Enrichment

- Several studies document changes due to enhanced Nitrogen deposition
- Effects are varied; empirical data
  - Array results using GIS to examine commonalities



# Western Nitrogen Enrichment





# Additional Effects

- We are evaluating how best to characterize additional effects.  
Qualitatively address:
  - Sulfur and Hg methylation
  - N<sub>2</sub>O on climate

Attachment G: 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  
Dr. Andrzej Bytnerowicz  
Ms. Lauraie Chestnut  
Dr. Ellis B. Cowling  
Dr. Douglas Crawford-Brown  
Dr. Charles T. Driscoll, Jr.  
Dr. Paul J. Hanson  
Dr. Dale Johnson  
Dr. Donna Kenski  
Dr. Naresh Kumar  
Dr. Myron Mitchell  
Mr. Richard Poirot  
Dr. Armistead Russell  
Mr. David Shaw  
Dr. Kathleen Weathers

## 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 NO<sub>2</sub> and SO<sub>2</sub>?*
- 2. How well characterized are the relevant properties of the ambient air concentrations and deposition of NO<sub>x</sub> and SO<sub>x</sub>, 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<sub>x</sub> and SO<sub>2</sub> chemistry, and its role in nitrification, and, *second* (and a distant second!), because its oxidation can be a minor source of NO<sub>x</sub>. 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 (O<sub>3</sub>): 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<sub>x</sub> 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 NO<sub>3</sub> 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 SO<sub>2</sub> and NO<sub>x</sub> 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 SO<sub>2</sub>, SO<sub>3</sub> though small in amount compared to SO<sub>2</sub> (about 1 to 3 percent of total emitted SO<sub>x</sub> 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 SO<sub>2</sub> 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<sub>x</sub> Emissions: Lines 14-18: The statement that sources other than electric utilities “make only a very minor contribution” to overall SO<sub>2</sub> emissions is quite inaccurate and needs to be corrected. In 2003, the non-EGU sources in the US contributed 31% of the total SO<sub>2</sub> 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 SO<sub>2</sub> 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 NH<sub>3</sub> 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<sub>x</sub>, NO<sub>x</sub>, 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 NO<sub>2</sub>: Say “NO<sub>2</sub> 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<sub>x</sub> and SO<sub>2</sub>” 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<sub>x</sub> and SO<sub>x</sub>: This is well-written section and does a very good job of responding to the Charge Question 2 on the subject of PRBs of NO<sub>2</sub>, SO<sub>2</sub>, 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](http://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.

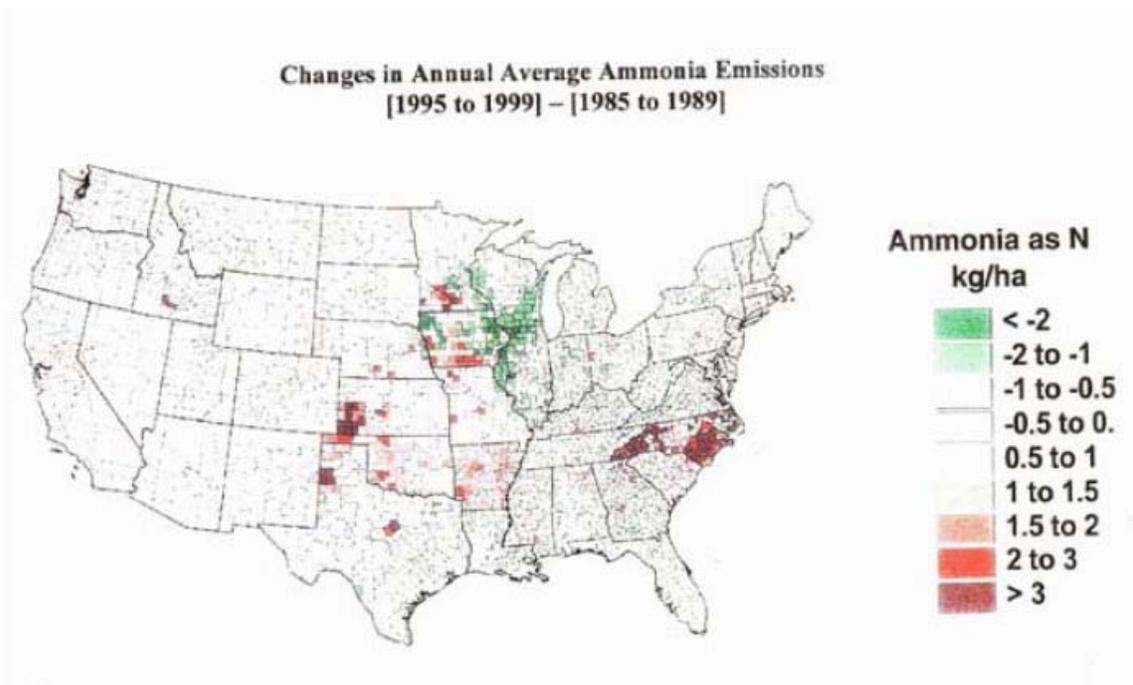
Page 5-3 Line 30: Please see the comments for Chapter 2 about SO<sub>2</sub> emissions from sources other than power plants. They are not “very little,” since they are about 1/3 of the total SO<sub>2</sub> emissions.

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<sub>x</sub> emissions in the eastern US because of the “NO<sub>x</sub> 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<sub>x</sub>, SO<sub>x</sub>, NH<sub>3</sub>, 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., NO<sub>2</sub>, SO<sub>2</sub>, HNO<sub>3</sub>, NH<sub>3</sub>, particulate NO<sub>3</sub>, particulate NH<sub>4</sub>).

Page 4-5, point 7 – ammonia (NH<sub>3</sub>) 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 NO<sub>2</sub> and SO<sub>2</sub>.

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 SO<sub>2</sub> on plants.

Pages 4-38 through 4-44. Change SO<sub>4</sub><sup>+</sup> to SO<sub>4</sub><sup>2-</sup>.

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 HNO<sub>3</sub> or of total dissolved NO<sub>3</sub><sup>-</sup>?

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 HNO<sub>3</sub> 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 HNO<sub>3</sub> toxicity than NO<sub>3</sub><sup>-</sup> deposition. There is also a high probability that the reduced N resulting from high NH<sub>3</sub> and particulate NH<sub>4</sub><sup>+</sup> 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 HNO<sub>3</sub> 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<sub>x</sub> as it contributes to nitrogen enrichment of the ecosystems appropriately identified and characterized?

Generally the effects of NO<sub>x</sub> 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 NH<sub>4</sub>NO<sub>3</sub> or (NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub> 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  $\text{NH}_3$ , 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  $\text{HNO}_3$ ,  $\text{NH}_3$  or particulate  $\text{NO}_3^-$  or  $\text{NH}_4^+$ , fertilization experiments may quite poorly mimic the real-world interactions between the atmosphere and ecosystems (vegetation, soils and surface water components).

### **Recommendations:**

1. While  $\text{SO}_2$  and  $\text{NO}_x$  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  $\text{SO}_4$  in wet precipitation measured nationally (e.g. NADP network). However, for N, not only wet  $\text{NO}_3$  and  $\text{NH}_4$  deposition (also measured nationally), but also dry deposition of gaseous  $\text{HNO}_3$ ,  $\text{NH}_3$  and particulate  $\text{NO}_3$  and  $\text{NH}_4$  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  $\text{HNO}_3$  and  $\text{NH}_3$  “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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**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 SO<sub>2</sub> and NO<sub>2</sub> 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 SO<sub>2</sub> and NO<sub>x</sub> 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 NO<sub>2</sub> and SO<sub>2</sub>?**

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<sub>x</sub>" and "SO<sub>x</sub>," 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<sub>x</sub> and SO<sub>x</sub>, 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<sub>x</sub> and SO<sub>x</sub> 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<sub>x</sub>, SO<sub>x</sub>, 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<sub>x</sub> and SO<sub>x</sub> 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<sub>x</sub> 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 N<sub>2</sub>O 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<sub>x</sub> 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 SO<sub>2</sub> 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<sub>x</sub> and SO<sub>x</sub> 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 NO<sub>3</sub>-. 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  $\text{NO}_x$  and  $\text{SO}_x$  (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  $\text{Cl}^-$  deposition.  $\text{Cl}^-$  is probably largely derived from coal combustion or industrial processes at least away from the coast. Both base cations are  $\text{Cl}^-$  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  $\text{NH}_3$  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.,  $\text{kg/ha-yr}$  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 <sup>+</sup>                                                                                                              |
| 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 <sub>x</sub> ] 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 <sub>2</sub> ]... 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 <sub>3</sub> alone or together with NO <sub>x</sub> 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 <sub>4</sub> <sup>+</sup> ) are provided.                                                                      |
| Page 3-2             | Section 3.1.2.1 Soils. This section needs some citations.                                                                                                       |
| Page 3-2, line 20    | Isn't N <sub>2</sub> the dominant soil gas?                                                                                                                     |
| Page 3-2, line 30    | Is there a typo here? Do you mean "Although N <sub>2</sub> O is not reactive...?"                                                                               |
| Page 3-3, line 4     | Do you mean...oxidation of NH <sub>3</sub> 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 <sub>2</sub> 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 <sub>2</sub> 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 ...<br><br>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 <sub>2</sub> ] ... 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 <sub>3</sub> /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 <sup>th</sup> 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 <sub>3</sub> . 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 <sub>x</sub> and SO <sub>x</sub>                                                                                                                                                                                                                                                                                      |

|                       |                                                                                                                                                                                                      |
|-----------------------|------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|
| Page 3-51, line 20    | Change to: The annual mean [NO <sub>2</sub> ] ...[NO <sub>2</sub> ] (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 <sub>2</sub> emissions.                                                                                                           |
| Page 3-59, line 29    | Again, that statement about the NH <sub>3</sub> 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 SO<sub>4</sub><sup>2-</sup>. 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 $\text{NO}_3^-$ are typically less than $\text{SO}_4^{2-}$ 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.<br>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<br>...                                                                                                                                                                           |
| Page 4-17, line 20    | Change to: ... Many streams that exhibit chemical conditions<br>during ...                                                                                                                                                                           |
| Page 4-17, line 24    | Need to define ANC                                                                                                                                                                                                                                   |
| Page 4-17, line 25    | Change to: in the central Appalachian Mountain region ... (a<br>reference is also needed to document this statement)                                                                                                                                 |
| Page 4-18             | Figure title. The figure is incorrect for the figure title and text<br>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<br>summer and spring ANC during baseflow conditions was on<br>average 30 µeq/L. This means that acidic episodes would occur on<br>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<br>moderate to severe episodic acidification during high flow that<br>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  $\text{Ca}^{2+}$  in the forest floor less important than a 50% loss of  $\text{Ca}^{2+}$  in the mineral soil?
- Page 4-25, line 2 Do we actually know that Al only becomes mobilized after  $\text{Ca}^{2+}$  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 $\text{NO}_3^-$ 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 $\text{NO}_3^-$ 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 $\text{Ca}^{2+}$ 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 $\text{H}^+$ 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...<br>(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 $\text{SO}_4^{2-}$ adsorption<br>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)...<br>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 $\text{SO}_4^{2-}$ is decreasing due to the long-term accumulation of $\text{SO}_4^{2-}$ 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: ... $\text{NO}_3^-$ 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 $\text{SO}_4^{2-}$ 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 $\text{SO}_4^{2-}$ ? 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 $\text{NO}_3^-$ plus $\text{NH}_4^+$ 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 <sub>4</sub> /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 $\text{SO}_4^{2-}$ 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 $\text{SO}_2$ . The effect of $\text{SO}_2$ 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 $\text{NO}_3^-$ 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 <sup>2+</sup> 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.<br>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).

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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<sub>x</sub> as an ozone precursor. NO<sub>x</sub>-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<sub>x</sub> 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<sub>x</sub> and SO<sub>x</sub> on plant response, but the quantitative understanding of the generally high ambient concentrations needed to illicit adverse responses to direct NO<sub>x</sub> or SO<sub>x</sub> 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<sup>-1</sup>y<sup>-1</sup> in the context of ambient levels that typically are maxed out near 10 kg N ha<sup>-1</sup>y<sup>-1</sup>.

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<sub>x</sub> and SO<sub>x</sub> 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 HNO<sub>3</sub> is incorporated into foliage:

- Hanson PJ, Garten CT (1992) Deposition of H<sup>15</sup>NO<sub>3</sub> 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 (HNO<sub>3</sub>) 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 NO<sub>2</sub> 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 (NO<sub>2</sub>)-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<sub>x</sub> by plants supplied with N-15-labelled fertilizer. *New Phytologist* 138:13-18.
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- 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 (NO<sub>2</sub>)-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) NO<sub>2</sub> 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<sub>x</sub> and SO<sub>x</sub> 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<sub>x</sub> concentrations through time (i.e., over decades). Is this data not available? The graphics provided for SO<sub>x</sub> 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<sub>x</sub> and SO<sub>x</sub>. 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<sub>x</sub> and SO<sub>x</sub> regulations. Delegating the primary effects of NO<sub>x</sub> and SO<sub>x</sub> 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 NO<sub>2</sub>-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 SO<sub>2</sub> and NO<sub>2</sub> 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 O<sub>3</sub>, NO<sub>2</sub>, their combination, and different soil N supplies. *CANADIAN JOURNAL OF FOREST RESEARCH* 26:649-657.
- Kainulainen P, Holopainen JK, Oksanen J (1995) Effects of SO<sub>2</sub> 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 SO<sub>2</sub> and NO<sub>2</sub>. *ENVIRONMENTAL POLLUTION* 107:421-436.
- MENG FR, COX RM, ARP PA (1994) Fumigating mature spruce branches with SO<sub>2</sub> 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 NO<sub>2</sub> by soybean leaves. *NEW PHYTOLOGIST* 136: 239-243.
- Qiao Z, Murray F (1998) The effects of NO<sub>2</sub> 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 NO<sub>2</sub> by spruce (*Picea-abies* L Karst) trees. I. NO<sub>2</sub> 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 SO<sub>2</sub> and NO<sub>2</sub> 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 SO<sub>2</sub> 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<sub>x</sub> 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<sub>x</sub> 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 CO<sub>2</sub> from the atmosphere. This could be added as a counter point to the discussion of N<sub>2</sub>O 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<sub>x</sub> and SO<sub>x</sub>

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 N<sub>2</sub>O as a greenhouse gas. How well does the draft ISA characterize the evidence on these topics?**

Designating the direct effects of gas phase NO<sub>x</sub> and SO<sub>x</sub> 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 N<sub>2</sub>O, the production of tropospheric ozone, the contributions of NO<sub>x</sub> and SO<sub>x</sub> 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 [CO<sub>2</sub>], 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 CO<sub>2</sub> 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 CO<sub>2</sub>-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 CO<sub>2</sub>."

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<sup>-1</sup>yr<sup>-1</sup> 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 CO<sub>2</sub> 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 CO<sub>2</sub> 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 CO<sub>2</sub>-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, Al<sup>3+</sup> as well as H<sup>+</sup>.

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: “SO<sub>4</sub><sup>+</sup>”? Since when is sulfate a cation? This is an embarrassing mistake. Sulfate is an anion: SO<sub>4</sub><sup>2-</sup>

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<sup>+</sup>, 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<sub>x</sub> 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<sub>x</sub> emissions. We're told that soil contributes 10% of NO<sub>x</sub> emissions globally, and 26% of NO in Illinois, but no estimates are given for soil NO<sub>x</sub> 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<sub>x</sub> emission trends, parallel to the discussion of SO<sub>x</sub> emission trends in Sec. 3.2. While changes in NO<sub>x</sub> have not been as dramatic as those for SO<sub>x</sub>, 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<sub>x</sub> and SO<sub>x</sub> sources. These analyses have been done by EPA already. Another significant hole in the emissions discussion was NH<sub>3</sub>. Although we're told that the NEI underestimated NH<sub>3</sub> 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<sub>x</sub> or SO<sub>x</sub> or NH<sub>3</sub>. I don't understand why the authors of this section decided to show us plots of ozone and PM<sub>2.5</sub> air quality but not plots of SO<sub>2</sub>, NO<sub>x</sub> or NH<sub>3</sub> concentrations. While it is true as stated in lines 11-13 that NO<sub>x</sub> and SO<sub>x</sub> are often monitored at the same sites, the number of NO<sub>x</sub> and SO<sub>x</sub> monitors is far less than for ozone or PM<sub>2.5</sub>. More relevant would be a plot of just NO<sub>2</sub> and SO<sub>2</sub> 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<sub>x</sub>/SO<sub>x</sub> 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<sub>x</sub> and SO<sub>x</sub> 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<sub>x</sub> and SO<sub>x</sub> 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<sub>x</sub> and SO<sub>x</sub> monitors are rare. How does the lack of rural NO<sub>x</sub> and SO<sub>x</sub> monitors affect estimates of N deposition?

Figures 3.7-2 and 3.7-3, which compares ambient SO<sub>2</sub> and SO<sub>4</sub> 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 SO<sub>4</sub> concentrations in particulates or in rainfall – its not clear which. A similar set of plots should be shown for NO<sub>x</sub> and NH<sub>3</sub>.

I didn't like Fig. 3.7-4 when it showed up in the SO<sub>x</sub> 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 SO<sub>2</sub> 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<sub>x</sub>, NH<sub>3</sub>, 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  $\text{NH}_x$  isn't addressed comprehensively here because the focus is  $\text{NO}_x$  and  $\text{SO}_x$ , and it may not be comprehensively discussed in the PM ISA, because the focus there is PM. When or where does  $\text{NH}_x$  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  $\text{NH}_x$ '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 ...25<sup>th</sup> and 75<sup>th</sup> 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,  $\text{SO}_4^{2-}$  morphed into  $\text{SO}_4^+$ . 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?

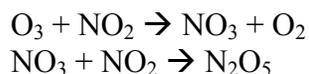
## Dr. Naresh Kumar

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<sub>x</sub> emissions from mobile sources and electric utilities (e.g., According to NEI 2003, 55% of NO<sub>x</sub> 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, NO<sub>3</sub> 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 SO<sub>4</sub><sup>2-</sup> 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 H<sub>2</sub>O<sub>2</sub>, O<sub>3</sub> or O<sub>2</sub> 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 O<sub>2</sub> 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 O<sub>2</sub>.

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: H<sub>2</sub>SO<sub>4</sub> should be changed to SO<sub>2</sub>.

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 NH<sub>4</sub>NO<sub>3</sub> from the list.

Page 2-26, Line 6: After “PM<sub>10</sub>”, add “, partly because NO<sub>3</sub><sup>-</sup> contributes smaller fraction to PM<sub>10</sub> and partly because NO<sub>3</sub><sup>-</sup> is present in a non-volatile form, such as NaNO<sub>3</sub>, in the coarse mode.”.

Page 3-1, Lines 23 & 24: Electrical utilities and various industries account for about one-third of anthropogenic NO<sub>x</sub> 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<sub>2.5</sub> 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<sub>Coarse</sub> mass, ions, and XRF elements;*
- 3. 24-hr gaseous ammonia as collected with an annular denuder;*
- 4. continuous (minute to hourly) PM<sub>2.5</sub> mass, OC, EC, ammonium, nitrate, and sulfate; light scattering and light absorption;*
- 5. continuous gaseous ozone, nitric oxide, nitrogen dioxide, total oxidized nitrogen (NO<sub>y</sub>), 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<sub>2.5</sub> 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<sub>x</sub> precursors, the factors controlling the
- 2-6 Change to: on the conventional 100-year time horizon of ~296 – i.e., N<sub>2</sub>O is nearly 300 times more effective on a per molecule basis for trapping heat in the atmosphere than carbon dioxide (CO<sub>2</sub>)(IPCC,
- 2-13 Indicate location of “Mace Head” in line 3.
- 2-18 Be more explicit in what is the interference by NO<sub>z</sub> compounds in line 1.
- 2-20 Reword to clarify the issue related to: also lead to artifact volatilization and associated positive bias in [HNO<sub>3</sub>] 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 NH<sub>3</sub> and NH<sub>4</sub><sup>+</sup> is NH<sub>x</sub>.)
- 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 N<sub>2</sub> is not reactive in the troposphere, N<sub>2</sub>O 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 (NH<sub>2</sub>) groups in amino acids.
- 3-3 Change to: will make emissions from this source increasingly important
- 3-8 This statement needs clarification: Emissions of SO<sub>2</sub> 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 SO<sub>2</sub>. 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 SO<sub>2</sub>?
- 3-9 Be more explicit about which pattern in the statement: Anthropogenic emissions of NH<sub>3</sub> show a strikingly different pattern from those of NO<sub>x</sub> or SO<sub>2</sub>.
- 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 comose 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 <sup>35</sup>S 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 NO<sub>3</sub>-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  $\text{SO}_4^+$  to:  $\text{SO}_4^{2-}$
- 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  $\text{NO}_3$  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  $\text{NO}_3$  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<sup>-1</sup> yr<sup>-1</sup>). 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<sup>-1</sup> yr<sup>-1</sup>, respectively) in the western Adirondacks than at HF (4.6 kg N ha<sup>-1</sup> yr<sup>-1</sup>) in the central Adirondacks. The greatest increase in nitrate loss in response to the experimental treatments occurred at HF where the HNO<sub>3</sub> 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<sub>x</sub>/NO<sub>x</sub> 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 – SO<sub>2</sub> or NO<sub>2</sub> – 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 1<sup>st</sup> 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 2<sup>nd</sup> half of chapter 2 seems unnecessarily limited to measurements of the “named” NO<sub>x</sub> and SO<sub>x</sub> 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<sub>x</sub> & NO<sub>x</sub> 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) HNO<sub>3</sub> on the list.

p. 2-3, Figure 2.2-1: Some inorganic nitrates – like NH<sub>4</sub>NO<sub>3</sub>– 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 Hg<sup>0</sup> to Hg<sup>2+</sup>, 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 NH<sub>4</sub>NO<sub>3</sub> 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 NO<sub>2</sub> are less soluble than SO<sub>2</sub>, but they are also much more reactive and, with their “longer-lived reservoir species HNNO<sub>3</sub>” do not necessarily transport over longer distances than SO<sub>2</sub> and its transformation products.

p. 2-17, lines 7, 9, 13, 14: Here it looks like you use “NO<sub>x</sub>” 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/NO<sub>2</sub> 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 NO<sub>3</sub><sup>-</sup> from (relatively acidic) PM<sub>2.5</sub> filters (if Teflon – not nylon) and more likely to be positive nitrate artifacts on alkaline PM<sub>10</sub> (or PM<sub>10-2.5</sub> filters), unless preceded by HNO<sub>3</sub> 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 NO<sub>2</sub>. See for example : [http://datafedwiki.wustl.edu/index.php/2007-07-18\\_ESIP\\_Demo\\_OMI\\_NO2](http://datafedwiki.wustl.edu/index.php/2007-07-18_ESIP_Demo_OMI_NO2) for a better illustration of & links to OMI NO<sub>2</sub> 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 SO<sub>4</sub>, NO<sub>3</sub>, Cl, or PM mass (which is irrelevant for TSP). Quartz filters can also have (smaller than glass but significant) positive artifacts from SO<sub>2</sub> or HNO<sub>3</sub>. Denuders are not really much of an option for hi-vol PM<sub>10</sub> samplers – on which quartz filters are typically run, but yes, denuders on lo-vol Teflon filter PM<sub>10</sub> or PM<sub>10-2.5</sub> (by dichot) sampling would help eliminate artifacts.

p. 2-26, lines 6-7: Actually, the loss of pNO<sub>3</sub> from FRM PM<sub>2.5</sub> 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<sub>2.5</sub> 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) HNO<sub>3</sub> & NH<sub>3</sub> concentrations in the indoor environment.

p. 2-27, lines 3-21: Why not move the discussion of negative pNH<sub>4</sub> artifacts adjacent to the discussion of positive pNH<sub>4</sub> artifacts beginning line 21, p. 2-25. Also I question your implication that negative artifacts generally exceed positive artifacts for NH<sub>4</sub>. Can you provide a reference for this? NH<sub>4</sub> is not measured in IMPROVE and NH<sub>3</sub> denuders are not used in STN – which does attempt to quantify NH<sub>4</sub>. 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 pSO<sub>4</sub> as parallel to pNO<sub>3</sub> in line 29.

p. 5-2, line 7” “controlling SO<sub>2</sub> oxidation” isn’t really the right term here. Could reword as something like “...altering the chemistry of SO<sub>2</sub> 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 NH<sub>3</sub>.

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<sup>-1</sup> 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  $\text{SO}_4^{2-}$  (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 “SO<sub>4</sub> 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 1<sup>st</sup> line under Nitrate: Add s to 1980. And 2<sup>nd</sup> 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<sub>x</sub> and SO<sub>x</sub>, 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<sub>x</sub> and SO<sub>x</sub>, 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 (SO<sub>2</sub> or SO<sub>3</sub>), 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  $\text{NO}_x$ ,  $\text{NH}_3$ ,  $\text{NH}_4^+$ ,  $\text{NO}_3^-$ ,  $\text{SO}_2$  and  $\text{SO}_4^{2-}$  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  $\text{pSO}_4$ ?

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  $\text{N}_2\text{O}_5$ . 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,  $\text{NO}_3$  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  $\text{NO}_3^-$  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  $\text{NH}_3$  measurement is needed.

2-27: Section 2.7.1: It is weird that  $\text{pNO}_3$  is included here,  $\text{pSO}_4$  is not.

2-28:28: There is a summary paragraph for  $\text{NH}_3$  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<sub>x</sub>, N<sub>2</sub>O, reduced N and SO<sub>x</sub> 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<sub>x</sub> 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<sub>x</sub>-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 NH<sub>3</sub>?

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 SO<sub>4</sub><sup>+</sup> instead of using SO<sub>4</sub><sup>=</sup> (or SO<sub>4</sub><sup>2-</sup>). 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<sub>x</sub>?

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<sub>x</sub> and SO<sub>x</sub> 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

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#### 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 SO<sub>2</sub> 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<sub>x</sub>. Include the primary standard for SO<sub>x</sub>. 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<sub>x</sub> and SO<sub>x</sub>.

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<sub>x</sub> and SO<sub>x</sub> omission**

The first paragraph states "The scope of the joint NO<sub>x</sub> and SO<sub>x</sub> 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<sub>x</sub> and SO<sub>x</sub> will be addressed within the secondary PM NAAQS." The omission of particulate phase NO<sub>x</sub> and SO<sub>x</sub> 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<sub>x</sub> and SO<sub>x</sub> 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 NO<sub>2</sub> and SO<sub>2</sub>?**

---

- 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<sub>x</sub> and SO<sub>x</sub>, 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 NO<sub>2</sub> and SO<sub>2</sub> to characterize ambient levels, but not enough information in rural areas and sensitive ecosystems.
- In general, emission inventories for SO<sub>2</sub>, whose sources are well-characterized, are more reliable than estimates for emissions of NO<sub>x</sub>, and certainly far more reliable than NH<sub>3</sub> emissions.
- There is more information, in terms of spatial coverage, on wet deposition of SO<sub>4</sub>, NO<sub>3</sub>, and NH<sub>4</sub> than ambient SO<sub>2</sub>, NO<sub>2</sub>, or NH<sub>3</sub>

#### **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<sub>x</sub> and NH<sub>x</sub>?

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<sub>x</sub> and NO<sub>x</sub> deposition on climate change. In addition, the effects of climate change on SO<sub>x</sub> and NO<sub>x</sub> pollution.

### **Adequacy of existing air monitoring network**

- The density of SO<sub>2</sub> and NO<sub>2</sub> monitors is adequate to track the primary standard, but is not adequate for the secondary standard.
- The bulk of the ambient SO<sub>2</sub> and NO<sub>2</sub> monitors, as well as the Speciation Trend Network for fine particulate SO<sub>4</sub> and NO<sub>3</sub>, are located in urban and suburban areas.

- There is no national NH<sub>3</sub> monitoring program, and what little information is available for ambient concentrations is research-oriented.
- There is information on fine particulate SO<sub>4</sub> and NO<sub>3</sub> 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 SO<sub>4</sub>, NO<sub>3</sub>, and NH<sub>4</sub> than there are ambient NO<sub>2</sub> 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<sub>y</sub> 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<sub>y</sub> 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<sub>x</sub> as a precursor**

NO<sub>x</sub> should be discussed as a precursor to the following:

- Ozone
- Ambient nitrates
- N deposition
- Fine particulates

### **Inclusion of N<sub>2</sub>O**

N<sub>2</sub>O is certainly a greenhouse gas, but is not a component of NO<sub>y</sub>, nor is it relevant to O<sub>3</sub>/PM/haze formation or N deposition (and subsequent acidification or nutrient enrichment). More information on particulate N would be favored over N<sub>2</sub>O 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<sub>x</sub>, NO<sub>x</sub> and NH<sub>x</sub>? For the primary and secondary standard? In general, emission inventories for SO<sub>2</sub>, whose sources are pretty well-characterized, are more reliable than estimates for emissions of NO<sub>x</sub>, and certainly far more reliable than NH<sub>3</sub> 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<sub>x</sub> and NO<sub>x</sub> 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<sub>x</sub> and SO<sub>x</sub> 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 HNO<sub>3</sub> wet and dry deposition, some areas of the country are experiencing a rise in NH<sub>3</sub> emissions and NH<sub>4</sub> 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 NH<sub>4</sub>-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<sub>x</sub> and SO<sub>x</sub> 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 NO<sub>3</sub> 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 SO<sub>2</sub> and NO<sub>x</sub> emissions and SO<sub>4</sub><sup>2-</sup> and NO<sub>3</sub><sup>-</sup> 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 <sup>15</sup>N: 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 <sup>15</sup>N 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].

Attachment H: Compilation of Individual Panel Member Comments on EPA's *Draft Scope and Methods Plan for Risk/Exposure Assessment: Secondary NAAQS Review for Oxides of Nitrogen and Oxides of Sulfur* (March 2008 Draft)

**Comments received:**

Dr. Praveen Amar  
Dr. Andrzej Bytnerowicz  
Ms. Lauraine Chestnut  
Dr. Ellis B. Cowling  
Dr. Douglas Crawford-Brown  
Dr. Charles T. Driscoll  
Dr. Paul J. Hanson  
Dr. Rudolf Husar  
Dr. Dale Johnson  
Dr. Donna Kenski  
Dr. Naresh Kumar  
Dr. Myron J. Mitchell  
Mr. Richard L. Poirot  
Mr. David J. Shaw  
Dr. Kathleen Weathers

## Dr. Praveen Amar

This write up provides comments in three areas, as requested by Dr. Ted Russell, the Panel Chair. These three areas relate to:

1. Adequacy of how well the REA (Risk/Exposure Assessment) covers some of the “additional” issues such as maple decline, ammonia air deposition, etc. The exact charge question #11 related to “additional effects” is reproduced here :

*Additional ecological/welfare effects due to NO<sub>x</sub> and SO<sub>x</sub> emissions that we do not currently anticipate evaluating in detail in this review include the following:*

- Nitrogen saturation,
- Maple decline,
- Ammonia air deposition and toxicity to native mussels,
- Relationship between acidity/nutrient enrichment and mercury methylation,
- Sensitive areas for acidity/nutrient enrichment impacts, identified from biogeochemical characteristics, and
- Climate change effects due to N<sub>2</sub>O.

*Does the Panel agree that these represent lower priority effects for the current assessment? If not, what does the Panel recommend?*

2. Sections 2.3.2 ( overview of nitrogen deposition) and Section 2.3.3 ( overview of sulfur oxides and sulfur deposition), and
3. Overall general comments on the complete Scope and Methods Document

First, a general comment about the whole document. I think it is generally written better than the ISA. However, various chapters can and should be improved for more clear communication. It is understandable that various sections have different authorships. These sections and chapters would therefore benefit from the services of an expert technical/scientific editor resulting in a more readable document that more clearly communicates what the proposed Risk/Exposure Assessment is expected to accomplish.

### **Charge Question # 11**

The Scope and Methods Document currently proposes to assess the areas mentioned above under Charge question #11 in a qualitative manner (Page 15; Appendix A.2, etc.). My sense is that some of these areas are of greater importance than others. For example, understanding of the relationship between sulfur deposition and enhanced methylmercury (MeHg) production in aquatic systems has serious policy implications as a number of mercury control programs from many industrial sources (coal-fired power plants, municipal waste combustors, steel mills, industrial boilers) are being put in place in the US at the same time major SO<sub>2</sub> reduction programs are being implemented (e.g., CAIR). To the extent sulfur deposition levels would be

changing in the US at the same time Hg deposition levels would be changing, this “integrated” assessment needs to be indeed “integrated.”

I do not have the expertise on the direct toxicity of ammonia on mussels, however, this Risk Assessment would benefit greatly from more emphasis on *all* science-related issues related to ammonia: better quantification of its emissions, its atmospheric chemistry and physics, its fate and transport, wet and dry deposition of ammonia and its products, and ecological effects. As other members of this CASAC Panel have noted in their comments, the ISA as well as the S and M document simply seem to address the role of ammonia as an afterthought and do not address the issue of ammonia at a level it deserves.

I suggest that the areas in Charge Question #11 (at least some of them) be given more attention than what is being proposed. For example, instead of addressing them in any future Risk/Exposure Assessment document in an Appendix, they be addressed in the body of the report itself as short and separate chapters. It is recognized that some of treatments might be more qualitative or empirical. That does not make them less important.

### **Sections 2.3.2 and 2.33 (Nitrogen and Sulfur Deposition):**

Sections 2.32 and 2.3.4 are short. I do not think they provide true “overview” of nitrogen deposition, or for that matter, sulfur deposition. Section 2.3.2 mostly covers the history of how N<sub>2</sub>O was addressed in previous standards review and then notes that it will not be covered in a quantitative way in the current assessment. It is important, as I note above, that all the “additional issues” including the role of N<sub>2</sub>O as a greenhouse gas be covered in this document in a clear and well-referenced manner (for example, the recent 2007 IPCC reports). One suggestion is that a revised Scope and Methods document remove these sections from their present location and integrate them more effectively with other sections.

### **Overall General Comments on the Document**

Page 1, Line 7: “... deposition of ambient particles, GASES, AND ACIDS, that can..”

Page 3, Line 12 “THOUGH” is an awkward way to start this sentence.

Page 5, Second Para: “welfare *effects* identified should be PROTECTED from adverse *effects*.” Need to rewrite.

Page 6, Section 2.2: Please be very careful when using the words *uncertainty* and *variability* in the same sentence unless you explain their clear and very different meanings. Many people take them to mean the same thing and they simply are very different concepts, both in science and in policy-setting.

Page 8, Last paragraph: “this assessment would focus on ecological quality and its effects from acidification...” This is the first and the only time this document uses the words “ecological quality.” What exactly is meant? Also, the sentence needs to be rewritten for clarity.

Page 9, Line 8, Please change to “precursors NO<sub>x</sub> and volatile organic COMPOUNDS...”

A General Comment: I would very much like to reinforce what Dr. Cowling noted in his comments on the September 2007 Draft Plan. These words speak more clearly than anything I have seen relative to what needs to be done in this assessment for the joint secondary standard(s) for revising/modifying/establishing a new/ standard(s):

- i) *“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?”*
- ii) *“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?”*

Page 11, Second Paragraph, “... Framework to analyze alternative *air quality standards* ...” Why not also include deposition standards as well as critical loads here? A quick review of this document also makes it clear that critical loads as an *organizing principle* for establishing standards need to be paid more attention (just like the role of ammonia needs to be paid more attention).

Page 24 (also other places): I suggest that this document be clearer about ecosystem services, valuation, valuation methods (both quantitative and non-quantitative). Since these areas are covered in the environmental economics and decision analysis areas, it would be a very good idea that they are described more clearly for the general audience of this assessment.

Page 25: The “Seven-Step Process” to undertake REA: I do not claim expertise, but Steps 5, 6, and 7 (“when feasible, scale up case studies...”, assess the current ecological conditions, assess alternative levels of protection...”, etc.) do not seem as rigorous as they would need to be to lead to a defensible standard(s).

Page 28: I am not so certain that ‘organic acids are common NATURAL sources of acidity in surface waters.’ I would note that many VOCs do lead to secondary organic aerosols (SOAs) and then to organic acids that are not “natural.”

## **Dr. Andrzej Bytnerowicz**

### General Comments

The document describes Problem Formulation, the first phase of the Ecological Risk Assessment (ERA) for ambient NO<sub>x</sub> and SO<sub>x</sub>, and focuses on the last component of the Problem Formulation Phase - the Analysis Plan. The document is logically structured and follows seven clearly described steps. The plan presents interesting ideas for development of the secondary standard for NO<sub>x</sub> and SO<sub>x</sub>. End points and conceptual models are also discussed but with much less detail.

In the beginning of the documents a clear statement is made that for evaluating ecological effects, total reactive N (both in the oxidized and reduced forms) will be considered. This is because reduced N compounds may be responsible for many effects caused by oxidized N. This is obvious and clear from the point of view of the present scientific knowledge. However, this is a long stretch from NO<sub>x</sub> (NO and NO<sub>2</sub>) and SO<sub>x</sub> which are the criteria pollutants for which secondary standards are being considered. I believe that legal considerations of such an expansion of the NO<sub>x</sub>/SO<sub>x</sub> term should be discussed upfront. If not, it may be expected that problems with various stakeholders during public discussion could take place.

Consideration for alternative secondary standards to ambient concentrations of NO<sub>x</sub> and SO<sub>x</sub> should also be addressed. For N, all forms of reactive N could be considered as part of the secondary standard. NO<sub>x</sub> (NO and NO<sub>2</sub>) is only a small portion of the problem, however, theoretically it is possible to link the secondary pollutants (such as HNO<sub>3</sub> or particulate NO<sub>3</sub>) to the primary pollutants (NO and NO<sub>2</sub>). Ammonia is the key pollutant for which concentrations and spatial and temporal distribution should be better characterized.

Considering high probability that relationships between NO<sub>2</sub> concentrations (or reactive N; Nr index) to N deposition, and between N deposition and its ecological effects may be weak, the deposition-based indices such as critical loads (CL), should be considered as a potential secondary standard. However, in order to consider such an approach a nation-wide monitoring of n deposition, similar to the European ICP Forests and ICP Vegetation, should be in place. Based on the existing monitoring efforts in the U.S., secondary standard for NO<sub>x</sub> and SO<sub>x</sub> based on N deposition could be considered only for a small portion of the country.

Options such as “cap and trade”, or emission reduction goals for specific administrative areas (states, counties, air quality management districts) should be addresses and considered. What was accomplished for the reductions of the SO<sub>2</sub> emissions in the Midwest and Northeast based on the requirements of Title IV of the CAAA or what is presently being done in Europe under the auspices of the Convention on Long-range Transboundary Air Pollution (CLRTAP) could be used as examples of the alternative approaches and possibilities (although such measures may be politically difficult to be accomplished). However, a note of caution – successful reduction of the SO<sub>2</sub> emissions was relatively easy considering that SO<sub>2</sub> sources are mainly large point emitters. For NO<sub>x</sub> such possibilities could be difficult to accomplish because of the complexity of the N chemistry and multiple sources of emissions.

Compared to the main ISA document, transitional ecosystems are not mentioned. For consistency this document should also treat them separately (especially since these ecosystems are very sensitive to the N inputs).

Diagrams used in the document greatly help a reader to visualize the planned risk assessment process envisioned by the authors. Figures 2-5, 3-1, or 3-5 are most useful.

A discussion of how the climatic conditions, or climate variability, could affect the proposed risk assessment process is lacking. Meteorological variability is mentioned, but this is not sufficient.

More emphasis should be placed on how the other co-occurring stressors (tropospheric ozone, drought, pests, diseases or catastrophic fires) affect N cycling and biological effects of N deposition.

Are the CMAQ estimates of N deposition updated? If I recall the CMAQ simulation for the western US, the latest model runs were done for 2002 (Gail Tonnessen, personal communication).

It would greatly help in review of this document if the lines were numbered!

### Specific comments

Page 1, first paragraph. A sentence “As discussed in the Integrated Review Plan (U.S. EPA, 2007a), this was done in recognition of the important linkages between ambient nitrogen and sulfur leading to deposition of ambient particles that can have significant impacts on environment”. This is only partially true considering that particles have only very small effects on the total N or S deposition. Gases or dissolved ions are much more important. Therefore, I suggest deleting “of ambient particles”.

Pages 7 through 9, Section 2.3. I found this section confusing and I suppose that other readers might have a similar experience. As I have stated in the beginning of my comments, this document is supposed to represent the Problem Formulation Phase. As such I would expect that the end points and conceptual models would get the same level of detail as the Analysis Plan. Although the conceptual models are described in section 2. “Background and Overview”, specifically in section 2.3.1, a discussion of endpoints is not easily found. Sections 2.3.2. “Overview of nitrogen deposition” and 2.3.3. “Overview sulfur oxides and sulfur deposition” are too short to present a good discussion of the problem. I suggest skipping them completely and referring a reader to the comprehensive review of these problems in the main ISA document.

Page 19, Table 2.1. The proposed schedule of the reviewing process is tight and very ambitious. The CASAC public review of the REA document in October is very early considering that the draft will be released in August. In my particular case, a very active schedule of conferences in August, September and October and summer vacation, does not leave enough time for a thorough review. Therefore, I recommend moving the CASAC review to November 2008.

Page 27, Table 3-1. For aluminum mobilization, terrestrial ecosystems (T) should be added. In the same table, for N concentrations, also terrestrial ecosystems are affected.

Page 31, 3.2, Step 2 (Define sensitive areas) and page 35, 3.3, Step 3 (Select risk/exposure cases study assessment area within a sensitive area) - use of natural gradients of N and S deposition should be considered.

Page 32, first two lines. I would argue that areas with good linkages to larger ecoregions should be also considered for new case studies. Results from such area could be extrapolated by means of geostatistics into much larger areas.

Page 35, last paragraph. I would like to suggest that “new N” could be called “reactive N species”, such as ammonia or nitric acid. “Old N” would be N stored in ecosystems as NO<sub>3</sub>-.

Page 38, first paragraph. Again – use of natural gradients for selection of the appropriate assessment areas should be considered.

Page 38, I would also suggest placing special emphasis on some areas in California, such as the San Bernardino Mountains, coastal sage and desert ecosystems that have been intensively studied and are sensitive to N additions.

Page 40, Table 3-6. Western states are under-represented. Please see my comment above.

Page 43, bullets. More appropriate would be using the term of “nitrate and ammonium deposition”. Ammonium would result from dry deposition of NH<sub>3</sub>, particulate NH<sub>4</sub><sup>+</sup> and water dissolved NH<sub>4</sub><sup>+</sup> deposition.

Page 57, first paragraph. After establishment of the baseline conditions, more advanced modeling techniques (dynamic models) would have to be used. Such models are able to take into account time effects and may be useful for ecological risk assessment. During the last few years such models have been used by the ICP Modeling and Maps in Europe.

The same paragraph. While particular loading “X” can cause a certain negative effect “Y”, it would be very difficult to translate it to ambient concentration of a pollutant (especially if NO and NO<sub>2</sub> are concerned). That effect “Y” would result from deposition of multiple N species. Is CMAQ, or other models, able to specify what concentrations of individual pollutants are responsible for the loading “X”? Maybe checking how the RAINS model used in Europe deals with such issues is necessary at this step of the analysis planning?

#### Comments on bullets from Page 57

“What adverse effects are we trying to protect against?” Best approach would be to look at multiple sensitive indicators (like sensitive lichens, C/N ratio, N foliar levels, mycorrhizae colonization). Use of single indicator could be misleading. However, for practical reasons and large-scale estimates, a single, easy to determine indicator (such as NO<sub>3</sub>- in soil solution) should be considered as a good initial step.

Next bullet – information on contribution of N oxidized and reduced forms to total N pollution (reactive N) effects is still poor. More intensive field campaigns and long term monitoring using some new techniques is urgently needed. A key problem is a need for better understanding of ammonia concentrations in the evaluated ecosystems.

Effects do occur due to different forms of N. Effects of gaseous N species (NO, NO<sub>2</sub>, NH<sub>3</sub> or HNO<sub>3</sub>) are very different among each other, and quite different from the effects of particulate or water-dissolved NO<sub>3</sub><sup>-</sup> or NH<sub>4</sub><sup>+</sup>.

How should alternative levels be selected? Combination of various methodologies is needed (controlled experiments, field exposures including natural gradients, others).

What are the correct temporal and spatial scales for the ambient air indicator? Ideally hourly values should be available. However, considering a lack of proper monitoring equipment that could be used at large scales, the weekly, bi-weekly or even monthly averages of the key deposition drivers would greatly help our abilities to model deposition. In such cases, passive samplers for gaseous pollutants could be used. Information on particulate component of dry deposition is poor, and its contribution to the total deposition may greatly vary in space and time.

Spatial coverage would depend on the chemical and topographic complexity of the area of interest. There are new geostatistical methodologies available that may help in optimizing monitoring networks and adequate spatial coverage.

Large-scale monitoring of throughfall could be used for integrative measure of wet and dry N deposition. However, use of the throughfall analysis methodology in arid and semi-arid ecosystems is questionable.

Comments on using 9 steps to determine the appropriate level and a form of a standard:

Generally this is a logical and well defined plan.

Point 1 – portion of dry deposited N, both in gaseous and aerosol forms, is not well characterized.

Point 2 – OK.

Point 3 – here, contribution of the main contributors to dry deposition of N, ammonia and nitric acid have to be better characterized.

Point 4 – the most sensitive indicators, and sets of various indicators, have to be considered. Although I generally agree with a need for determining ecosystem services and bundling them in order to find comparisons between locations, I am not sure if that would not complicate the establishment of a link between effects, deposition, ambient concentrations and emissions.

Point 5 – OK.

Point 6 – I suggest considering collaboration with the European scientists and modelers experienced in using RAINS model. Impacts of meteorological (climatic) variability are of a very high importance.

Point 7 – critical loads concept would be very useful.

Point 8 – intuitively I agree. However, I have to get acquainted better with the available literature to say more.

Point 9 – OK.

#### Responses to the assigned questions:

Question 8. In the seven-step approach to the current conditions risk/exposure assessment, Step 5 (Section 3.5) discusses how to scale up case study areas to more spatially extensive sensitive areas, where appropriate. Does the Panel agree with this approach or can they suggest alternatives?

The proposed approach seems to make sense as long as the extrapolations are done within the same ecoregions. However, distribution of concentrations of the key drivers of N dry deposition, specifically nitric acid vapor and ammonia, may dramatically change between the urban and agricultural sources and the receptor areas due to their high reactivity and deposition to vegetation and other surfaces. Therefore the on-ground determinations of patterns of their distribution are recommended. Passive samplers offer a relatively simple and reliable method for determining pollution concentrations. Results of such monitoring efforts aided by geostatistics may be used for generation of concentration surfaces of these pollutants. For estimates of dry deposition of these gases inferential methods could be used at large geographic scales. In addition, complexity of soils characteristics have to be also taken into account in proposed scaling-up exercises.

Question 9. In the seven-step approach to the current conditions risk/exposure assessment, Step 6 (Section 3.6) outlines a path to assess the current conditions of sensitive ecosystems. How well does the Panel agree with the approach outlined for calculating response curves and utilizing mapping and ecosystem services to characterize current conditions or can the Panel recommend alternative approaches?

This is a valid approach. Response curves from controlled experiments or natural gradient studies encompassing wide ranges of deposition levels would provide databases needed for estimates of current conditions and models simulating future changes. Critical loads (CL) calculations (performed with steady state, empirical or dynamic models) have been used in the NE USA and Canada for many years. In Europe, CL have also been successfully measured on the ICP Forests and ICP Vegetation networks. The ICP Modeling and Maps converts that information to maps representing annual levels of CL and exceedances for N and S deposition and acidity. The European Regional Air Pollution Information and Simulation (RAINS), uses those results for controlling of the internationally agreed quotas of emissions for individual countries. A similar approach could be tested and possibly used in the U.S. as well. Instead of

individual countries (as in Europe), states or air quality management districts could be used as the administrative units of control.

- a. How well does the Panel agree with using ecosystem services to provide a common metric for comparing ecological risks due to nitrogen and sulfur deposition effects?

This could be very useful approach for integrating effects of deposition of N and S on ecosystems. By presenting examples of what services are affected, potential stakeholders would be able to better comprehend implications of the adverse effects of elevated N and S deposition. That could bring more attention and potential support from the general public, NGOs and other stakeholders which would help in development of the secondary, welfare effects-based standard.

- b. How well does the Panel agree with collecting current valuation studies to understand the value of bundled ecosystem services? Can the Panel recommend additional or alternative approaches?

I generally agree with this approach. A holistic approach to the variously affected services seems to be logical. However, it should be evaluated if these new complex attributes of changes would not complicate the entire process of secondary standards development. Maybe as the first step towards development of a new standard the well tested European approach (CL and RAINS combined) should be initially considered. The idea of ecosystems services (including their bundling) could be considered as the next step leading to further improvement of the standards.

Question 10. In the seven-step approach to the current conditions risk/exposure assessment, Step 7 (Section 3.7) describes an approach to assess degrees of protection/levels of effects under alternative forms and levels of ambient NO<sub>x</sub> and SO<sub>x</sub> standards. This approach attempts to describe how the methods, models, and results of the current conditions risk/exposure assessment can inform our evaluation of the appropriate form(s) and levels(s) of a national standard. How well does the Panel agree with the approach outlined in this section, the issues presented, and the 9 steps outlined to assess potential forms and levels of the standard? Please suggest any additional or alternative steps we should take into consideration?

The section presents an interesting plan for defining and describing ecological changes caused by N and S deposition and an attempt to associate them with the NO<sub>x</sub> and SO<sub>x</sub> concentration-based standards. Although not easy to implement on a national scale, the critical loads (CL) methodologies could be used for defining levels of the acceptable deposition of N, S and acidity. Maps of CL exceedances, as it has been done for the NE United States, eastern Canada and Europe, can be very useful in showing the areas where the unacceptable deposition has taken place. Such information aided by appropriate models could lead to recommendations of where the N deposition reductions are needed in order to prevent deterioration of sensitive ecosystems. While it may be possible to use such information for recommending emission reductions, as it is being done in Europe under the Convention on Long-Range Transport of Air Pollution (please see my remarks above), a direct link to ambient NO<sub>x</sub> or SO<sub>2</sub> concentrations would be extremely difficult, if not impossible, to establish.

Plan presented in Section 3.7 establishes a framework of the logically aligned steps (activities) that theoretically could lead to development of new, concentration-based standards for NO<sub>x</sub> and SO<sub>x</sub>.

## **Ms. Lauraine Chestnut**

One general comment: There will undoubtedly be limitations in data for this assessment. I want to recommend that there be an effort throughout the process to define data that will improve this assessment the next time around. Collection of some of these data may be ongoing, but there may not yet be enough to answer the quantitative questions: the need for these monitoring and data collection efforts to have continued support should be articulated. There may be further monitoring and data collection that would be helpful. These need to be defined and prioritized so that resources are put to their best use and support is generated for more monitoring and data collection with focus on the specific information needed to help answer the key policy questions.

### Section 2.4

1. It seems really important for this assessment to address the question of whether both S and N have to be reduced to obtain benefits, or are they causing separate effects that are essentially additive? This speaks to whether the standards for the two need to be linked.

2. On page 18, I don't understand the question about whether long-term effects and cumulative loadings are relevant to the NAAQS review. If they are important for understanding the effects on ecosystems, why would they not be relevant?

### Charge question 4:

The need to define and describe the nature of the harm to the ecological resource that might be considered an adverse effect needs to be considered right at the beginning when the indicators are reviewed and selected. It is important that the selected indicators be able to be linked to changes in deposition, but also to changes in ecosystem services. There may be a threshold or a continuous relationship between levels of the indicators and harmful effects on ecosystem services (or even a nonlinear relationship that entails positive effects over some ranges and negative effects over other ranges). Either way, it is important to ask whether a given change in the selected indicator can be interpreted as showing a harmful effect, the implications of which can be articulated for the policy maker. For example, do we know at what level for each indicator the ecosystem loses capacity to support some species that would normally be expected to live in this system? It will be very important to be able to explain why the change in the indicator matters and what the significance of the change is in terms of ecosystem functions and services.

### Charge question 9b:

Once the losses or impairments in ecosystem functions and services at current deposition rates are quantified (or otherwise characterized) then it will be useful to search for available valuation studies that provide relevant information. It does not seem useful to try to determine total values for all ecosystem services even in the selected case study areas, because the policy relevant valuation issue for these pollutants is probably not a total loss in services, but a marginal change in quality or quantity of some services. To the extent that the literature supports valuation of bundled ecosystem services it makes sense to use this approach, especially if these services are

perceived as connected or joint products of the ecosystem functions, and if there are multiple services affected by the same change in pollution exposure.

Charge question 11:

I think EPA has selected the right categories of effects to focus the quantitative risk assessment efforts. This list of other effects needs to be included and described. In some cases the assessment might provide partial quantitative information. For example, will it be possible to say what the expected changes in ambient concentrations of N<sub>2</sub>O might be expected as a result of alternative standards? I agree it is not appropriate to expend resources here to try to quantify the associated climate change effects.

## **Dr. Ellis B. Cowling**

### **Individual Comments on the Draft Scope and Methods Plan for the Risk/Exposure Assessment: Secondary NAAQS Review for Oxides of Nitrogen and Oxides of Sulfur**

Before plunging directly into my specific assignments to critically review the “Key Policy Relevant Questions” in Section 2.4 and answer Charge Questions 3-7 in preparation for the April 3, 2008 CASAC Consultation, let me emphasize how pleased I was to see in this Draft Scope and Methods Plan how much EPA appears to be progressing in its willingness to think in a more holistic way about:

- 1) Multiple pollutants and multiple effects of airborne pollutants that often occur in the same air parcel and frequently act together to cause similar, different, and/or sometimes more pronounced effects than single pollutants acting alone.
- 2) Broadening the “Indicator of Concern” for the secondary NAAQS for “Oxides of Nitrogen” to include chemically reduced and organic forms as well as oxidized forms of “total reactive nitrogen,”
- 3) Recognizing that NAAQS designed to protect ecosystems from adverse effects of air pollutants often require different pollutant indicators, levels, averaging times, and statistical forms than NAAQS designed to protect human health.
- 4) The multiple similarities and advantages rather than just the differences among “NAAQS,” “Critical Loads” and “Target Loads” for nitrogen and sulfur air pollutants.
- 5) How “the words we choose often reveal the quality (and sometimes also the limitations) in our understanding” about various phenomena in nature. Useful examples in the current Draft Plan include the significance of such words as “ecosystem services;” “acidification,” “nutrient enrichment,” “nitrogen saturation,” “NO<sub>x</sub>/SO<sub>x</sub>,” “NO<sub>x</sub>/NH<sub>x</sub>/SO<sub>x</sub>,” “total reactive nitrogen,” “acid precipitation,” “acidifying deposition,” “wet plus dry acidifying deposition,” Etc.

Please note a few additional comments below regarding some of these important matters and especially the Resolution displayed on page 3 of these Individual Comments after it was passed by the Integrated Nitrogen Committee of the EPA Science Advisory Board for Communication on October 31, 2007 to our CASAC NAAQS Review Panel on the Secondary Standards for Oxides of Nitrogen and Oxides of Sulfur.

**My Specific Assignments in this CASAC Consultation on the Draft Scope and Methods Plan for the Risk/Exposure Assessment Secondary NAAQS Review for Nitrogen Dioxide and Sulfur Dioxide**

My specific assignments in preparation for our April 3, 2008 CASAC Consultation on the Draft Scope and Methods Plan as outlined in Chairman Ted Russell's memo of 11 March 2008 are:

- 1) Key Policy Relevant Issues. These topics are summarized in Section 2.4 in the Draft Scope and Methods Plan, and
- 2) Charge questions 3-7 – Ecological Effects Characterization. These Charge Questions are listed in Lydia Wegman's memo to Kyndall Barry dated 5 March 2008.

Chairman Russell also gave the first of these two assignments to six other CASAC panel colleagues – Douglas Crawford-Brown, Paul Hanson, Naresh Kumar, Myron Mitchell, Kathleen Weathers and Lauraine Chestnut. He also gave the second assignment to three other CASAC Panel members – Charles Driscoll, Myron Mitchell, and David Shaw. Thus, I am very much looking forward to comparing notes with all of these other people during our Consultation on April 3<sup>rd</sup>!

**Assignment 1) -- Critical Review of section 2.4:“Key Policy Relevant Questions”**

Both the current Chair of CASAC, Dr. Rogene Henderson, and others among the Statutory Members of our Clean Air Science Advisory Committee have written in recent months about the Need for Policy Relevancy as the Dominant Concern in the New NAAQS Review Processes.

In these carefully structured review processes, the most critical and overarching policy question that needs to be answered is:

*“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?”*

And similarly, especially with regard to the present integrated [simultaneous] review of the Secondary Standards for NO<sub>x</sub> and SO<sub>x</sub>, the most critical and overarching Key Policy Question is:

*“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.”*

With the latter overarching Key Policy Question in mind, we now have the considerable advantage that a much more complete focus can be achieved on **critically important scientific findings** that are **as directly relevant as possible** to this overarching Key Policy Question. To achieve this more complete focus, it is desirable that each chapter of the **Integrated Science Assessment**, the **Risk/Exposure Assessment**, and the **Policy Assessment/Rule Making** documents for each criteria pollutant should contain a summary section composed almost entirely of **very carefully crafted summary statements of conclusions and/or scientific findings**. These carefully crafted summary statements should:

- 1) **Contain the distilled essence of the most important topics covered in each chapter, and**
- 2) **Be as directly relevant as possible to the overarching Key Policy Question written above..**

Section 2.4 of the present *Draft Scope and Methods Plan* contains a series of 17 policy relevant questions to which carefully crafted scientific answers could be developed. But these mostly policy relevant questions need to be better organized and coordinated so that the answers to these questions can be used to build a coherent series of linkages between the major **acidification and nutrient enrichment effects of concern** and the **nitrogen and sulfur secondary NAAQS standards** that will be necessary to avoid these ecological effects.

My most serious reservation about the 17 policy relevant questions listed on pages 17 and 18 of Section 2.4 is that chemically reduced forms of nitrogen (NH<sub>x</sub>), organic forms of nitrogen (NC<sub>x</sub>), and total reactive nitrogen (TN<sub>r</sub>) all are not included in either the specific wording or the written context of any of these 17 questions. Only the first sub-part of the sixth question listed on page 17 deals explicitly with ammonia, ammonium ion, and/or total reactive nitrogen as alternative “indicators” of nitrogen pollution.

“Should the current standards for NO<sub>2</sub> (as an indicator for NO<sub>x</sub>) and SO<sub>2</sub> (as an indicator for SO<sub>x</sub>) be retained, revised, or revoked and/or replaced with alternative standard(s) having different indicators to provide the required protection from known or anticipated adverse welfare effects?”

I hope that these 17 policy relevant questions can be adjusted, revised, reorganized, and/or augmented in order to further increase their value in optimizing the quality of **summary statements of scientific findings** that will be as relevant as possible to the overarching Key Policy Question printed above in bold italic type.

In this connection, please also note once again the attached

**Resolution from the Integrated Nitrogen Committee of the Science Advisory Board for Consideration by the CASAC Secondary NAAQS NO<sub>x</sub> and SO<sub>x</sub> Review Panel**

During the ongoing meeting of the EPA Science Advisory Board’s Integrated Nitrogen Committee (INC) -- meeting at SAB Headquarters in Washington DC on October 29-31, 2007 -- the several members and Chair of the INC, Dr. James Galloway of the University of Virginia,

asked me (as the CASAC-designated liaison person to the Science Advisory Board's Integrated Nitrogen Committee) to present the following Resolution (which was developed and approved by the INC) for consideration during the CASAC review of the NAAQS for NO<sub>x</sub> and SO<sub>x</sub> during our CASAC Conference Call Consultation on October 30, 2007.

## **Resolution**

The current air pollution indicator for oxides of nitrogen, NO<sub>x</sub>, is an inadequate measure of reactive nitrogen in the atmospheric environment. The SAB's Integrated Nitrogen Committee recommends that inorganic reduced nitrogen (ammonia plus ammonium) and total oxidized nitrogen, NO<sub>y</sub>, be monitored as indicators of total chemically reactive nitrogen.

### **Assignment 2 – Charge Questions from OAQPS**

My comments are organized below in response to each of the several Charge Questions posed in Lydia Wegman's memo to Kyndall Barry dated 8 March 2008. As you will see much more detailed attention has been given to Charge Questions 3-7, to which Chairman Ted Russell asked me to give special attention.

- 1. In outlining the scope of this risk/exposure assessment, we have created a flow diagram that represents how nitrogen and sulfur compounds move from 'source to dose' in the environment (see Figure 2-1). How adequately does this conceptual model for evaluating risks due to deposition-related ecological effects characterize what should be covered in the scope of this assessment?**

This conceptual model seems very adequate as an initial frame of reference for deposition-related ecological risks

- 2. The main ecosystem effects areas we anticipate evaluating in this risk/exposure assessment are:**
  - (1) risks to terrestrial ecosystems from nitrogen enrichment effects,**
  - (2) risks to aquatic ecosystems from nitrogen enrichment effects (eutrophication),**
  - (3) risks to terrestrial ecosystems from acidification effects (nitrogen and sulfur), and**
  - (4) risks to aquatic ecosystems from acidification effects (nitrogen and sulfur).**

**We also plan to qualitatively discuss the role of sulfur enrichment on methylmercury production and the role of nitrous oxide in climate change. What key effects areas, if any, have been overlooked by this approach? Should the assessment plan be modified to include other effects?**

Especially considering the court ordered time constraints under which this Risk/Exposure Assessment must be completed, it seems very reasonable to concentrate on these the four major aspects -- nutrient enrichment and acidification in terrestrial and aquatic ecosystems but to also include some assessment of the other two effects of sulfur enrichment on methylmercury production and N<sub>2</sub>O influences on climate change.

- 3. Due to the complexity of conducting a nationwide risk/exposure assessment for welfare effects due to NO<sub>x</sub> and SO<sub>x</sub>, we have outlined a strategy designed to identify sensitive ecosystems and a range of harmful/adverse effects (see Figure 3-1). The seven steps are to**
- (1) identify documented biological, chemical and ecological effects and potential ecosystem services,**
  - (2) define sensitive areas using GIS mapping,**
  - (3) select risk/exposure case study assessment areas,**
  - (4) evaluate current loads and effects in case study assessment areas,**
  - (5) scale up the case study assessment areas to larger sensitive areas where feasible,**
  - (6) assess current ecological conditions in those areas, and**
  - (7) assess alternative levels of protection under different ambient scenarios.**

**Does the Panel agree with this general approach? Should it be improved or modified?**

Yes, I agree that this general approach is sound, But it certainly can be improved and modified in its presentation and implementation within the Risk/Exposure Assessment for which this is a Draft Plan.

As indicated on page 2 of these Individual Comments, my major reservation about the general approach outlined in this Draft Scope and Methods Plan is that it remains unclear if EPA is really willing and interested to include reduced (especially ammonia and ammonium) and organic forms of nitrogen (amino acids and volatile amines) as well as oxidized forms of nitrogen in the crucially important “indicators” being considered in this integrated (simultaneous) review of the present “NO<sub>x</sub>” and “SO<sub>x</sub>” NAAQS secondary standards.

It was reassuring to read in several parts of this Draft Scope and Methods Plan:(for example) on page 22 that:

“The risk exposure assessment will focus on ecosystem welfare effects that result from the deposition of total reactive nitrogen “ and also on page 26 that:

“To assess the impacts of total reactive nitrogen and sulfur loading we plan to identify adverse terrestrial and aquatic effects ...”

But it was also discouraging to see that the schematic diagram chosen for inclusion on page 12 of this Draft Scope and Methods Plan deals only with oxidized forms of nitrogen and includes no mention or pictorial illustrations of the huge (mostly agricultural) sources of reduced and organic forms of reactive nitrogen.

If EPA is serious about dealing with both “nutrient enrichment” and “acidification” of ecosystems induced by atmospheric deposition of total reactive nitrogen, the agency will have to learn how to sustain its recent success in learning to think more holistically (in part by learning how to write more holistically) about ammonia, ammonium ion, NH<sub>x</sub>, and NC<sub>x</sub>, and TN<sub>r</sub> about “ecosystem services” “critical loads,” and “multiple pollutant/multiple effects” approaches in air quality management.

What a different impression this Draft Scope and Methods Plan would have created if:

- the more inclusive abbreviation “NO<sub>x</sub>, NH<sub>x</sub>, and SO<sub>x</sub>) had been used instead of just “NO<sub>x</sub> and SO<sub>x</sub>,” or
- the words total reactive nitrogen” and “total nitrogen loading” were used more frequently and more appropriately:

- In formulating the Key Policy Relevant Questions listed on pages 17 and 18,
- In the captions of the tables and figures on pages 12, 16, 19, 28, and 29, as well as
- In preparing the major subheadings and topic sentences of paragraphs on pages 7, 15, 16, 19, 21, 22, 27, 49,55,56, 59, and A-3.

**4. In the seven-step approach to the current conditions risk/exposure assessment, Step 1 (Section 3.1) describes an approach to identify the documented effects, biological, chemical and ecological indicators, and potential ecosystem services related to acidification and nutrient enrichment. Does the Panel agree with this approach or can they suggest alternative approaches we should consider?**

I think you did very well in devising the seven step approach presented in Figure 3.1 -- the first step of which is described more fully in the text in Section 3.1.1 on pages 26-30. Thus I encourage your continuing use of this first step approach so long as you keep in mind that reduced and organic forms of nitrogen may be even more important than oxidized forms of nitrogen in many of the sensitive areas to which this approach is intended to be applied.

**5. In the seven-step approach to the current conditions risk/exposure assessment, Step 2 (Section 3.2) outlines a path to define areas sensitive to total reactive nitrogen and sulfur inputs. Do the Panel members agree with this approach or are there better alternatives that should be considered?**

Once again, I think your step 2 approach makes sense. I was especially pleased to see that your intention is to use GIS mapping as a tool to further analyze and interpret the strengths and limitations of similarities and differences among these selected sensitive areas.

**a. We are attempting to characterize the risks to ecosystems from sulfur and nitrogen deposition nationwide by clustering sensitive ecosystems where possible and by using the linkages between these areas at different scales. Please comment on the adequacy of this approach.**

I have only very limited personal experience in using these kinds of clustering analytical tools, thus I am not well qualified to assess the adequacy of these methods of analysis.

**b. How appropriate are the datasets and GIS maps listed in Table 3-4 for identifying ecosystems sensitive to nitrogen and sulfur and/or are there others that have been overlooked?**

The array of general types of ecosystems listed in Table 3-4 seems very adequate to me except for the lack of near-coastal oceans waters.

**6. In the seven-step approach to the current conditions risk/exposure assessment, Step 3 (Section 3.3) outlines a path to identifying risk/exposure case study assessment areas. a. Table 3-5 provides an initial list of indicators, mapping layers and multimedia models that may be used to assess ecosystem risk and exposure.**

**a. Please comment on the appropriateness of these and suggest alternatives that may be better suited for this analysis.**

Over the past 20 years, I have had some interactions with the principal investigators involved in about half of these studies of ecosystem effects, possible indicators, and the mapping layers and modules used in these kinds of terrestrial and aquatic problem areas. From these interactions, I have the general impression that these problem areas are appropriate for use in Step 3 and I have no suggestions of other methods that would be more suitable for this kind of analysis.

**b. Please comment on the list of potential case study assessment areas in Table 3-6 and Table 3-7 and make recommendations or suggest any alternatives.**

As in my response to the issues raised in Charge Question 6a, I have had some interactions with the principal investigators involved in about half of these studies and believe that the array of possible case study areas listed in Tables 3-6 and 3-7 is reasonable and is likely to be fruitful for the purposes of this Risk/Exposure Assessment and evaluation of benefits that are likely to occur from decreases in atmospheric loading of total reactive nitrogen. My experience is much more limited with respect to sulfur and mercury loading.

**7. In the seven-step approach to the current conditions risk/exposure assessment, Step 4 (Section 3.4) outlines a path to assess current nitrogen and sulfur loads and their effects on case study assessment areas. Does the Panel agree with how we have described our approach to identifying datasets, gaps, and uncertainties?**

The task of identifying useful data sets is challenging but the associate tasks of identifying gaps and characterizing uncertainties is even more complicated and challenging. The approach you have planned to use seems very sensible. I am also particularly pleased that you plan to stretch even further and attempt to characterize uncertainties in the context of current understanding of total reactive nitrogen impacts on ecosystem services rather than the more familiar measures of ecosystem growth, stability, and productivity.

**a. We have initially identified the primary chemical indicator that is most suitable for assessing ecosystem acidification effects as acid neutralizing capacity (ANC), with alternatives depending on data availability (see section 3.4.1 and Appendix B). Does the Panel agree with this selection, or can they suggest alternative/additional key indicators?**

I presume that you mean that atmospheric-deposition induced losses in ANC” rather than just ANC itself is “the most suitable chemical indicator for assessing ecosystem acidification effects. With this small adjustment in the phrasing of your question, I agree that ANC is probably the indicator of choice for assessing acidification effects in aquatic ecosystems. I know of no indicator that would be more suitable although maybe loss of cation exchange capacity might be worth some consideration. Charles Driscoll will be better able to answer this question more authoritatively than I am able to do.

**b. We have described the models being considered for this analysis (see section 3.4.2 and Appendix C). Does the Panel agree with the choice of these models, and can they help prioritize them for modeling the responses of the indicators recommended in Step 1 (Section 3.1)?**

I have no experience on which to base an informed judgment about the comparative merits of these models.

**8. In the seven-step approach to the current conditions risk/exposure assessment, Step 5 (Section 3.5) discusses how to scale up case study areas to more spatially extensive sensitive areas, where appropriate. Does the Panel agree with this approach or can they suggest alternatives?**

I have no experience on which to base an informed judgment in response to this question.

**9. In the seven-step approach to the current conditions risk/exposure assessment, Step 6 (Section 3.6) outlines a path to assess the current conditions of sensitive ecosystems. How well does the Panel agree with the approach outlined for calculating response curves and utilizing mapping and ecosystem services to characterize current conditions or can the Panel recommend alternative approaches?**

I agree that the approach outlined in Section 3.6 is appropriate for your assessment of the current condition of ecosystems. As indicated in my response to Charge Question 7 (above) I am very pleased

that you are planning to base your assessment on the magnitude of atmospheric deposition impacts on ecosystem services.

**a. How well does the Panel agree with using ecosystem services to provide a common metric for comparing ecological risks due to nitrogen and sulfur deposition effects?**

Once again, I am very pleased that you are planning to use ecosystem services as the base for your assessment of comparative risk due to total reactive nitrogen and sulfur deposition effects.

**b. How well does the Panel agree with collecting current valuation studies to understand the value of bundled ecosystem services? Can the Panel recommend additional or alternative approaches?**

I am not sure what is meant by “bundled ecosystem services” thus I have no basis on which to offer an informed judgment in answer to this question,

**10. In the seven-step approach to the current conditions risk/exposure assessment, Step 7 (Section 3.7) describes an approach to assess degrees of protection/levels of effects under alternative forms and levels of ambient NO<sub>x</sub> and SO<sub>x</sub> standards. This approach attempts to describe how the methods, models, and results of the current conditions risk/exposure assessment can inform our evaluation of the appropriate form(s) and level(s) of a national standard. How well does the Panel agree with the approach outlined in this section, the issues presented, and the 9 steps outlined to assess potential forms and levels of the standard? Please suggest any additional or alternative steps we should take into consideration.**

I am very pleased to see that you plan to do what appears to me to be a very thorough and effective analysis of alternative forms and levels of ambient “NO<sub>x</sub> and SO<sub>x</sub>” standards ( by which I presume you also mean alternative “NO<sub>x</sub>, NH<sub>x</sub>, NC<sub>x</sub>” or “total reactive nitrogen” standards” ).

The nine step approach you have outlined seems very sensible to me although I could not figure out how step 1 and 3 are different from each other, I was also puzzled about why you included the “levels and forms” of alternative standards and not also the associated “averaging time and statistical form” of alternative standards. Maybe the latter two aspects are so much less important than the first two that the latter two are not essential for the purpose of this analysis.

**11. Additional ecological/welfare effects due to NO<sub>x</sub> and SO<sub>x</sub> emissions that we do not currently anticipate evaluating in detail in this review include the following:**

- Nitrogen saturation,
- Maple decline,
- Ammonia air deposition and toxicity to native mussels,
- Relationships between acidity/nutrient enrichment and mercury methylation,
- Sensitive areas for acidity/nutrient enrichment impacts, identified from biogeochemical characteristics, and
- Climate change effects due to N<sub>2</sub>O.

**Does the Panel agree that these represent lower priority effects for the current assessment? If not, what does the Panel recommend?**

I agree that all of these effects are of lesser importance than the four main topics that are planned for inclusion – nutrient enrichment and acidification effects on both terrestrial and aquatic ecosystems.

But would encourage some reconsideration of both “nitrogen saturation” -- which is hard to avoid since it is the “limiting case” when you are already dealing with nitrogen enrichment effects.

I feel similarly about keeping climate change effects caused by N<sub>2</sub>O emissions in the present assessment plan --mainly because climate change effects are so likely to become very much more important in the future of our country's concerns about human induced changes in the chemical and physical climates of our planet.

## **Dr. Douglas Crawford-Brown**

This review focuses primarily on the Risk Assessment Framework presented, both in the Overview sections and in the step-by-step procedures described in Chapter 3 and in the appendices. The goal is to ensure both that the risk assessment goals are appropriate for standard-setting, and that the methods to be used will provide the kinds of information, of sufficient quality, to meet those goals.

The authors have focused appropriately on acidification and nutrient enrichment in the general framework for deposition-related effects from both S and N oxides, as these are the effects for which the scientific database is sufficiently sound. The decision to include perturbations on methylmercury also appears sound. My primary concern on all of these measures is whether loading, rather than subsequent concentrations, will be sufficient to accurately determine effects levels. I realize the data are primarily in the form of loadings, and so this may be unavoidable. But it still seems to me that there can be large variations in the relationship between loading and concentration due to differences in hydrological characteristics and chemical reaction rates (e.g. buffering) between water systems. But so long as the resulting variability and uncertainty can be characterized, this should not prove a large problem for establishing a NAAQS.

With respect to N<sub>2</sub>O as a greenhouse gas, I appreciate the attempt to place oxides of nitrogen into a framework that includes consideration of climate change, but can find little in the document to support the idea that it will prove possible to relate emissions to either GHG forcings or resulting changes in climate or ecosystem measures. This is an interesting idea that probably is not yet ready for NAAQS. It was wise, therefore, to leave the discussion qualitative on this point.

I remain unclear as to the role of the case studies. The decision to focus on especially sensitive ecosystems is a good one so long as it can be assumed that they are representative of a reasonably large class of such sensitive sites on the national scale (I don't think it would be appropriate to base national standards on situations at 1 or 2 outlier sites). So, the question that will naturally arise is what these case study sites are taken to represent in a national distribution. It would be good if some indication could be given of the number of such places in the US that might be equally sensitive and equally exposed (or some combination of sensitivity and exposure that makes them experience impacts at least as large as the case studies).

I realize that much of the work will depend on what is found in preliminary assessments of the data, but I was not at all clear as to how the spatial resolution was to be treated. Where there is high spatial variability, the exposure side of the exposure-response relationship can be greatly affected by choice of location. The authors do mention that they will explore this issue with examination of spatial gradients, but don't provide much detail as to how these gradients will be quantified to determine the appropriate grid size for atmospheric concentration, loadings and effects. Care will need to be taken in this aspect of the assessment.

The authors mention at one point that they will search for ways to “consolidate effects” effects across acidification and nutrient enrichment. It would be nice to have such a summary measure, but I doubt it will be possible to accomplish this goal. Instead, one will be left with a variety of what are essentially hazard quotients with no way to combine these into a summary hazard index. This won’t prevent setting a secondary NAAQS, however.

Figure 3-1 is a good summary of the steps to be taken. I don’t, however, know what Step 5 means (I understand it conceptually, but don’t know how it will be accomplished other than through pure judgment).

I am supportive of Step 1 in general, although the authors will need to think carefully about how they will ensure that decisions are based on adverse effects and not simply effects. As written, the document makes it sound as if any change in any indicator is adverse, an assumption with which I disagree. That can in part be dealt with by relating indicator changes to alterations in ecosystem services, although (i) the data to support such relationships are lacking and (ii) it still will be important to define what is meant by a significant decline (and not simply a change) in an ecosystem service).

While the use of GIS to examine the issue of ecosystem sensitivity may prove useful, I see nothing here that convinces me such a GIS-based study can provide fundamental insights into the causes of sensitivity. And I don’t fully understand why the GIS studies are needed. I would assume the causes of sensitivity are elucidated in the scientific literature, negating the need for a scientific study as part of the NAAQS process. The mapping layers selected appear appropriate, but again I can’t see how they will be overlain and assessed to either identify sensitive areas that can be used as case studies or to elucidate conditions that lead to sensitivity. But perhaps the authors will find a way to perform a useful assessment once they data are collected, visualized and analyzed spatially.

At one point, on Page 38, the authors comment that one wants to select a region where atmospheric effects dominate (at least, this is what I take them to mean). That is good for a scientific study of the effects of deposition, but a region can be sensitive to deposition precisely because there are other sources of nutrients that push the system close to some natural limits. So I am not sure that this criterion would be appropriate for selecting case studies.

The candidate assessment areas starting on Page 39 represent a good cross-section of potential case study areas. They will need to be narrowed to make the study feasible.

I have little expertise on modeling deposition, and so will not comment on that aspect. I note, however, that many of the models go well beyond modeling deposition, looking at speciation, concentration in water, etc. I was never sure which aspects of a particular model the authors were proposing to use, given the early parts of the document suggested that loading would be the exposure measure in exposure-response relationships.

Step 5 remains very “black box” to me. I think the idea is good, but can’t see the methodology that is being proposed.

Step 6 appears to be a form of hazard quotient, or margin of exposure approach. I support this in general as it is the best that can be done.

The natural services valuation will be tricky. There are significant conceptual problems with approaches that begin to mimic Costanza-like valuations, in large part because there is no real market in which these services are traded. So the values are hypothetical, in the sense of a kind of revealed preference. I suspect there will be a lot of disagreement on this aspect of the assessment.

If the framework in the first 6 steps can be worked out, then Step 7 should be relatively straightforward and I support the approach defined here.

## Dr. Charles T. Driscoll

### Comments on Draft Scope and Methods Plan for Risk/Exposure Assessment: Secondary NAAQS for Oxides of Nitrogen and Oxides of Sulfur

#### General and specific comments:

- Page 10 Does figure 2-1 imply that species alterations, eutrophication and mercury methylation are by-products of nutrient enrichment or are they relevant to both disturbances? Is there some significance to the fact that these are sub-bullets under nutrient enrichment? Please clarify.
- Page 14 2.3.4. Isn't nutrient enrichment (terrestrial) and eutrophication (aquatic) really the same thing? Why give these separate designations? This seems to confuse the issue.
- Page 15 Last paragraph. Why are sulfur-mercury interactions exacerbated in coastal waters more than other ecosystems? Is there any scientific basis for this statement?
- Page 16 Figure 2-5. Would it be useful to separate freshwater and coastal systems for aquatic effects? These are very different systems.
- Page 16 There is limited scientific evidence that increased  $\text{NO}_3^-$  leaching inhibits methylmercury production.
- Page 17 Bullet 4. This bullet is very focused compared to the other bullets. Could it be modified to make it more consistent with the other bullets? Something like "What is total sulfur and nitrogen deposition to ecosystems and how do values vary across the landscape?"
- Page 17 Bullet 6, sub bullet 2. I'm not sure I understand the context of this question: Does it refer to whether the effects of atmospheric nitrate deposition can be distinguished from effects associated with other inputs of nitrogen? This is a strange question. Can it be made more explicit?
- Page 27 Some of the indicators on the table don't make sense. What is aluminum or metal mobilization? How do you quantify or characterize forest health? Why are ANC and alkalinity both listed? Under nutrient enrichment should SAV density or hypolimnetic DO be listed? Should methyl or total mercury be included?
- Page 28 For acidification there should be a primary indicator of soil calcium status, maybe soil % BS, in addition to ANC which is indicative of surface water.

- Page 28 Why isn't deposition an indicator for acidification like nutrient enrichment?
- Page 28 Third line. Should this be carbon to nitrogen ratio?
- Page 28 Table 3-3. SAV area should be included.
- Page 45 Again, it is critical to have both a soil and surface water indicator for acidification. For example in the Northeast surface water ANC is increasing while soils are continuing to lose exchangeable  $\text{Ca}^{2+}$  even under current decreasing deposition. Because soil and surface waters are not completely coupled, two indicators should be used: maybe ANC and soil % BS.
- Page 47 If CMAQ is to be used to make deposition projections, it is critical that an effort be made to validate simulations of total and dry deposition. Models like MAGIC or PnET-BGC require historical (dating back to 1850) and future values of total deposition. Will CMAQ be able to do this? A data fusion exercise will be needed. It would be helpful to evaluate the compatibility of output data from CMAQ with the input requirements for the watershed models. This is a critical issue that will need to be addressed in a rigorous way.
- Page 48 Paragraph 3. Define GUI.
- Page 49 Is it possible to summarize this text by developing a table which lists the potential models, their characteristics, potential for application and limitations? It would be good to indicate which of these models have been used in exercises similar to what is proposed. My sense is that many of these models have never been applied to anything even close to what is proposed here. Also which of the models have been tested at any of the proposed study regions in a manner similar to what is proposed here? Have any of these models been linked with CMAQ and how successful has this been? Also, analysis of sulfate effects on mercury methylation was mentioned in the introductory materials. Are there any models available to assess this effect?
- Page 50 Paragraph 1. Note PnET-BGC can be applied in a shorter than monthly time-step, if that is desired. Most past simulations have been done in a monthly time-step to reduce computational time in long-term simulations.

Generally, the draft S&M Plan seems appropriate. That said, what is discussed is very general and not very specific. Also, what is proposed is very ambitious. I would imagine it would take considerable time and resources to conduct the proposed analysis. Little text was focused on critical loads. How does the Draft S&M Plan incorporate existing critical load efforts or is compatible with critical load efforts or plans?

## Dr. Paul J. Hanson

The plan contains appropriate general principals, but the presentation and terminology could be improved as outlined in the following comments. Policy relevant background levels of NO<sub>x</sub> and SO<sub>x</sub> exposures, acidification, and N & S deposition should be discussed or planned to be discussed somewhere in this document (see page A-8 for an exception). Adding line numbers to this document would have facilitated its review.

### Specific comments:

I would add definitions for the following terms used in the documents to the “Key Terms” pages:

- Adverse Effect/Change – See possible definitions in text on page E-1.
- Criteria pollutant
- Dose – see use on page 10.
- Ecological quality – used on page 8
- Indicator – This term is used two ways in this document. On the one hand it is used (I think correctly) in terms of the definition of a possible Secondary NAAQS. In other places it is used to describe a measure of change driven by NO<sub>x</sub> and SO<sub>x</sub> pollutants. The authors need to choose one form of use and stick with it throughout the document.
- Multimedia Modeling – Why isn’t this just ecological or process modeling?
- Sensitive Ecosystem

Page vii: Within the definition of Total Reactive Nitrogen the last line might include the phrase ‘N-containing organic compounds’.

Page 2 (top of the page): I’m not convinced that the authors should conclude that a more robust database will be available after another 5-year cycle. Does EPA have plans to fund significant new research? Are other agencies or organizations planning investments in N and S deposition effects research?

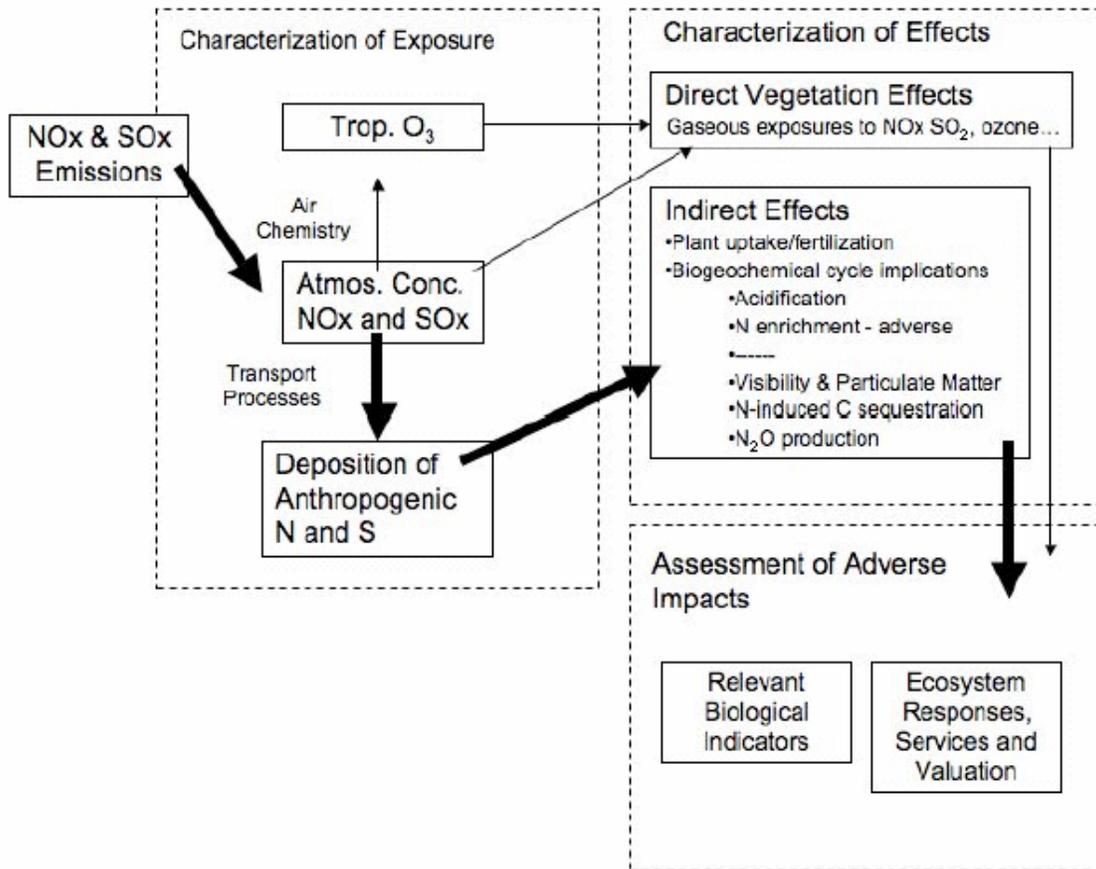
Page 2 (middle): The regulatory authority for addressing NO<sub>x</sub> and SO<sub>x</sub> as criteria pollutants is well described. More detail on how this authority translates to the subsequent extended discussion on N and S deposition effects (acidification, N enrichment) would be appropriate. A brief comment to this issue is added at the top of page 3.

Page 6: The previous NO<sub>x</sub> ACQD published in 1995 was not limited to a discussion of ambient air effects of NO<sub>2</sub> on vegetation. It included a large amount of data summarized from the acid rain literature on N deposition effects (or potential effects).

Page 8: The connection between NO<sub>x</sub> emissions, subsequent deposition and the direct generation of N<sub>2</sub>O solely from the anthropogenic N inputs is not strong. Should it really be included here? References or direction to the ISA could also be used at the bottom of page 8.

Page 9 second paragraph: The primary effect of NO<sub>x</sub> and SO<sub>x</sub> would be a direct effect on vegetation function/survival or a direct impact on a material surface. I would classify all of the effects discussed in this plan as secondary. The N and S deposition effects are at least one step removed from the form of the primary pollutant. After rewording, however, I would retain this paragraph. For complete appreciation of the impacts of NO<sub>x</sub> and SO<sub>x</sub> on public welfare, their contribution to the generation of atmospheric particles and tropospheric ozone is worth mentioning.

Figure 2-1 may reflect the proposed assessment process, but it doesn't illustrate the complexity of the problem in my opinion. I recommend something like the following alternate figure. In this example the bold arrows reflect the dominant processes to be focused on within the REA.



Page 11 4<sup>th</sup> line from the bottom: Should the word “reduced” be oxidized?

Top of page 12: The word “or” might be changed to ‘and’.

Page 15: In the first bullet, Risks should probably be associated with ‘adverse nitrogen enrichment effects’.

Figure 2-5: This is a much better figure than Figure 2-1. It might be moved forward and used together with Figure 2-1. It might also include other dashed ovals for additional known effects of

NO<sub>x</sub> and SO<sub>x</sub> that are simply not dealt with in this document (e.g., tropospheric ozone and aerosols, visibility, particulate matter).

The bottom full paragraph on page 16 needs a reference.

Bullets on page 17:

- 1 Good
- 2 May not be enough information
- 3 Good
- 4 Good
- 5 Critical
- 6 Current standards provide little protection nor do they inform the phenomenon of acidification or N enrichment highlighted in this document.
- 7 It isn't clear how anthropogenic NO<sub>x</sub> from atmospheric pollutant sources can be distinguished from other forms of N inputs (i.e., agricultural fertilizer).

Bottom of page 17: Ecosystem characteristics will be critical. This is a key issue.

Page 18: The bullet questions and key issues outlined on this page are very good. At the bottom of the page it may not be clear what is intended in the discussion of long-term impacts (weeks, months, years, decades??). A statement on the need to understand background levels of N and S deposition and natural processes of acidification in a regulatory context should probably be included.

Page 22: In the third paragraph 2nd line, please eliminate the word adverse. Not all effects are adverse.

Section 3.1.1: To avoid confusion with the use of the word 'indicator' as a term to describe the nature of the regulated pollutant (e.g., NO<sub>x</sub> and SO<sub>x</sub> atmospheric concentrations), I would change the use of indicator throughout this section to something like 'measures of response/change'.

Page 29: As a measure of nutrient enrichment, throughfall N deposition (Table 3-3) will fail to measure the component of rainfall N absorbed by the canopy. I favor the use of total N deposition as an indicator of pollutant exposure.

Page 30: A measure of ecological or biological change resulting from acidification or N enrichment will be difficult to isolate from trends in other biogeochemical cycling drivers (temperature, soil water status, herbivory outbreaks...).

Middle of page 35: The suggestions of the EES are well thought out and should be the basis for this evaluation. It will be critical to show that the anthropogenic source of the driver for adverse ecological changes is the cause for any perceived adverse effects. Can this be done?

Bottom of page 35: Can new and old N really be distinguished (or need to be distinguished) in the context of the N and S biogeochemical cycles? Distinguishing anthropogenic contributions to

N & S deposition might be accomplished via the use of stable isotope studies or measurements, but such observations will not be available for EPA's analysis in this REA.

Page 45: As an indicator of the pollutant I would favor combined wet and dry N deposition.

Using throughfall would underestimate total N additions (i.e., the canopy is a good filter for the extraction of N from rainfall).

Page 47: Of the monitoring networks mentioned on this page, several seem to be unrelated to air pollution (e.g., agricultural runoff modeling). They may be needed to calculate total N enrichment effects, but they will not inform air quality issues themselves.

Page 47: Add a reference for the "National Atlas".

Bottom of Page 55: The point made at the bottom of this page is really important. I don't, however, know how one will define a national standard specific to regional areas.

Page 57: Application of modeling for the purposes proposed by EPA in this document tends to assume that the models have validated mechanisms that 'get it right'. This may not actually be the case. EPA should be careful not to apply models validated for one area/region to other areas without appropriate attention to their utility in those regions. The authors should provide a more detailed discussion of the mechanisms needed by 'useful' models ahead of their description of the models chosen for the REA analyses.

Page 58 Bullet #5: Are sufficient data actually available to support the development of these relationships.

Page 58 Bullet #7: Where the effects from loadings are anticipated to accumulate over long time periods (i.e., years), it may be important to begin to also evaluate the potential implications of climate change on such relationships.

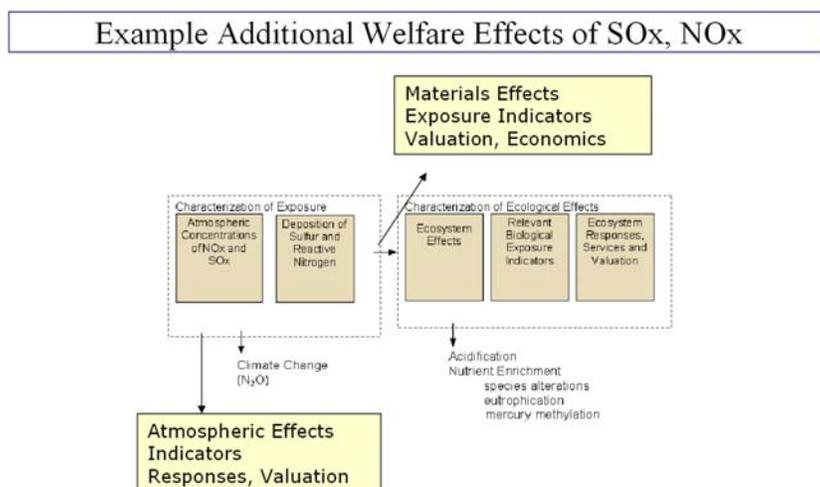
## Dr. Rudolf Husar

Overall, the draft plan, Scope and Methods section is for the Secondary NAAQS for NO<sub>x</sub> and SO<sub>x</sub> is thorough and executable. In particular, expanding the scope by combining NO<sub>x</sub> and SO<sub>x</sub> is commendable. The combined review plan is consistent with the intertwined and multifaceted nature of welfare impacts. However, there are several serious concerns regarding some aspects of the Scope and Methods sections:

### Question 1: Conceptual Model of the Welfare Risk

#### Consideration of other Welfare Effects of NO<sub>x</sub>/SO<sub>x</sub>.

The primary NAAQS is focused on protecting human health, while NAAQ Standards are to protect and minimize the **welfare** effects of pollutions. The planned scope of the NAAQS review document review is focused almost exclusively on the ecological effects of SO<sub>x</sub> & NO<sub>x</sub>. The known welfare effects such as perturbations to the atmosphere (weather, climate, optics), and the economic/esthetic effects on man-made materials are not covered.



Marginalizing these non-ecological welfare effects will omit a significant rationale for mitigating SO<sub>x</sub>/NO<sub>x</sub> emissions. This omission is also inconsistent with the recent trend toward integrated assessment of interlinked pollutant systems: While SO<sub>x</sub> and NO<sub>x</sub> are considered as a combined set of interacting pollutants, their combined effects are treated incompletely in piecemeal fashion. This limits the ability of the document to provide a full rationale for acting on welfare effects.

It is recognized that some discussion of welfare effects of SO<sub>x</sub>/NO<sub>x</sub> have been incorporated in the NAAQS Criteria document for PM. If deemed desirable, this document could make references to the specific sub-sections of the PM Document. However, the key findings relevant to the welfare effects should be present in this document, so as to provide a complete

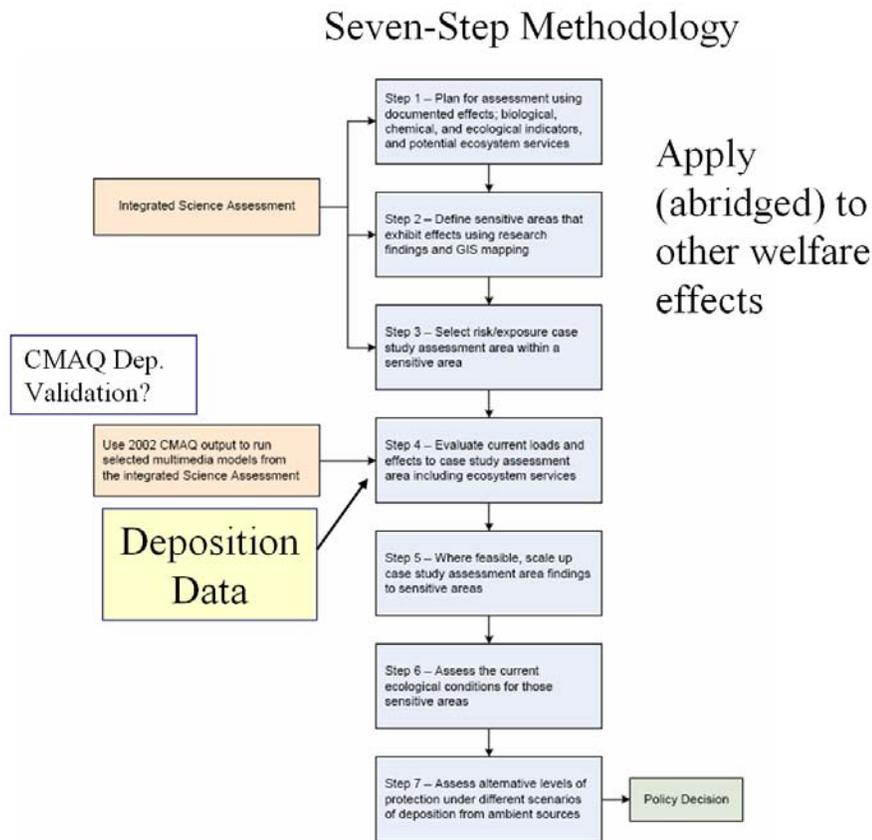
assessment. Also, as stated below, most other documents do not treat the question of indicators adequately.

The discussion at the RTP meeting has partially resolved my concern. A concise summary of the sulfate and nitrate – related visibility effects in this document still seems appropriate.

## Question 2: Seven-Step Methodology

### Apply Abridged 7 Steps to Non-Ecological Welfare Effects

The seven-step methodology proposed for the ecological effects is sound and executable. The same methodology, suitably abridged for each welfare effect should be applied to the other welfare effects. This would give consistency and scientific robustness to the document.



Elaborate versions of Steps 3, and 5 may not be necessary for some effects. However, the development of sensitive areas, indices, current conditions would be desirable for these welfare effects.

### 2.3.2 Overview of Nitrogen Deposition

This section gives appropriate general exposition of NO<sub>x</sub> chemistry, concentrations and deposition. In this overview, (including in Fig 2.2) it would have been helpful to indicate that

- NO<sub>x</sub> deposition is primarily through gaseous HNO<sub>3</sub>
- Total deposition is dominated by dry deposition

The addition of the NO<sub>x</sub> flow diagram discussed at the CASAC meeting will reduce my concern. Such a diagram should indicate the relative magnitudes of the wet/dry deposition processes.

The inclusion of N<sub>2</sub>O in this scoping document discussion is puzzling. It is stated that N<sub>2</sub>O is a greenhouse gas but also that. “Since that is outside the scope of this review, it will not be a quantitative part of this assessment”. So, how was the scope defined? What is the rationale for omission?

### 2.3.3 Overview of Sulfur Oxides and Sulfur Deposition

This section gives appropriate general exposition of SO<sub>x</sub> chemistry, concentrations and deposition. It is not clear what is meant by “Due to known acute effects on plants, in previous NAAQS reviews, SO<sub>2</sub> served as the chemical indicator for SO<sub>x</sub> species.” For the reviews over the past 30+ years, SO<sub>2</sub> and SO<sub>4</sub> have been clearly separated when presenting their concentrations and deposition. In what context was SO<sub>2</sub> a surrogate?

Pertaining sections 2.3.2 and 2.3.3, there is a general comment regarding the use of the CMAQ model. The heavy reliance on CMAQ model for (concentration and) deposition estimation is of concern. The CMAQ model performance for Sox is documented and for ambient SO<sub>4</sub> concentrations, it seems adequate. However, the marginal performance of CMAQ for Sox wet deposition and for non-sulfur species suggests considerable room for improvement. Additional effort directed toward CMAQ validation should be included. The direct use of observational data in estimating deposition (Step 4) and for extrapolating the case study areas (Step 5) should be considered.

The discussion at the CASAC meeting regarding CMAQ and its calibration/evaluation for use in this document has not eliminated my concern. Evidently, the CMAQ evaluation is still someone else’s problem...

### 3.2.2 GIS Mapping

This activity is commendable. This section outlines a very ambitious plan to generate many of ‘GIS layers’ for use in the assessment. In particular, **Table 3-5** summarizes the current plan for GIS mapping layers and models to be applied in the risk/exposure assessment of targeted sensitive ecosystems. These layers originate from a variety of sources and they represent very diverse content. The preparation of each layer and the integration of the layers to support the seven steps is a technically and scientifically challenging task.

It would appear useful to prioritize the preparation of data layers, test the layer- integration methodologies on simple data fusion analyses. Finally, the credibility of this would be enhanced if the GIS data layers prepared for the Review would be documented, cataloged, published and made available for re-evaluation and re-use by the regulatory and scientific community.

The EPA presentation at the CASAC meeting has addressed these issues adequately.

## Dr. Dale Johnson

### General Comments

As in my last two reviews, I stress the need for looking at all points of view, especially with respect to nitrogen deposition. As in the other documents, the focus is on the negative effects, which leaves this document open for severe criticism once it is released. This is especially true for the potential effects of nitrogen deposition on carbon sequestration in US forests – this could in fact be a negative effect of reducing N emissions! Once 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.

Question 4. How well are the major effects of NO<sub>x</sub> and SO<sub>x</sub> 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?

The effects of NO<sub>x</sub> and SO<sub>x</sub> on ecological acidification are fairly well characterized – that is not the problem. The problem is the scope of this assessment – namely, the focus on acidification. Nitrogen is the limiting nutrient for most terrestrial ecosystems and therefore there is the distinct possibility of beneficial effects as well. I believe that it is both a scientific and strategic mistake to ignore the potential benefits of N deposition to the national C budget while at the same time considering only one part of the greenhouse situation by including the negative effects of N<sub>2</sub>O. I daresay that climate change is potentially a much larger environmental issue than acidic deposition or N saturation is – at least it certainly looms much larger in the media and the mind of the public these days. Nadelhoffer et al. (1999) dismiss the potential benefits of N deposition on forest growth largely on the assumption that most deposited N is immobilized by microbial uptake in the forest floor and soil and only a small proportion (20% in their studies) is recovered by trees. This is in fact typical of previous forest fertilization studies which show that microbes typically out compete trees for N. However, the argument is disputed by Jenkinson et al (1999) find their argument unconvincing because it ignores foliar uptake: most atmospherically-deposited N first impinges on the foliage, not the forest floor, as in fertilizer studies and in the studies cited by Nadelhoffer et al (1999), and microbial competition in the forest canopy is considerably less than in the soil. At a minimum, the arguments by Nadelhoffer et al (1999) imply that greater rates of N deposition would lead to greater rates of forest C sequestration, an argument also implied by the analysis of Hungate et al (2003). Finally, a recent study by Pregitzer et al (2008) has conclusively demonstrated that experimental inputs of N at relatively low levels (30 kg ha<sup>-1</sup> yr<sup>-1</sup>) caused significant increases in C sequestration in live trees and forest floor in forests in Michigan.

Again, I emphasize that the point of delving into this issue in some detail is not to justify increases in N deposition but to fully ventilate both sides of that issue. The potential contribution of N deposition, especially at higher levels, to N deficient forests has not been resolved and needs some experimentation and careful analysis. The current proposed methods include only those

sites where N deposition has proven to have negative effects or is likely to have negative effects, and these sites are simply not typical of forests in the United States which are mostly N-limited rather than N saturated. Furthermore, the results from these arguably unusual N-saturated sites, which do not reflect the normal N limited condition of forests in the US, will then be extrapolated, apparently to represent N effects over the entire country. This is bias in the extreme – case studies need also to include chronically N deficient sites where growth and indeed even ecosystem health might improve with increased N deposition! I believe that such an analysis should be a part of the plan for assessing the effects of atmospheric deposition in the document being reviewed. It makes no sense to partition out the acidification effects of air pollution from the potential effects of C sequestration.

5. How well has the ISA characterized the relationship between acidifying deposition levels of NO<sub>x</sub> and SO<sub>x</sub> and environmental effects?

All of the comments made above apply to this question as well. The ISA has some technical problems in the way acidification is handled, as noted in my review, but the major problem is the complete focus on acidification and the lack of perspective on the larger C budget and global climate issues looming ahead.

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

There is some lack of clarity on this in the ISA document, as noted in my detailed review on p. 4-12, lines 2-9. Ammonium is acidifying whether it nitrifies or is taken up by plants or microbes (in the latter case, causing the release of H<sup>+</sup> during uptake). The oxidized form of N, nitrate, is not in itself acidifying – in fact, uptake of nitrate should cause the release of OH<sup>-</sup> to maintain charge balance in the plant or microbe.

#### Specific Comments:

p. 2. paragraph 2: Effects on climate are mentioned here – such effects must include the potential increases in C sequestration associated with N deposition from pollution in order to be a complete picture.

p. 14, last paragraph: “can be characterized as a positive or negative effects...” Only negative effects are sought in the case studies proposed in this document. No mention is made of potential positive effects on the bullet list on p. 15.

p. 26, top: “we plan to identify adverse terrestrial and aquatic effects..” Why only adverse effects? Positive effects were mentioned earlier – was this only lip service? Where is the balance?

p. 26, bottom: “focused on what soil features should be tracked, including how organic matter affects microbial processes.” I do not understand what is meant here by how organic matter affects microbial processes nor how it relates to the problem at hand.

p. 27, Table 3-1: Carbon budget, growth, etc are highlighted here – but this will apparently be assessed only in sites specifically selected to show the most adverse effects in the case study assessment. Furthermore, the results from these arguably unusual N-saturated sites, which do not reflect the normal N limited condition of forests in the US, will then be extrapolated, apparently to represent N effects over the entire country. This is bias in the extreme – case studies need also to include chronically N deficient sites where growth and indeed even ecosystem health might improve with increased N deposition!

p. 30, paragraph 1: among ecosystem services, the example of primary production is given. See comments directly above. Assessment of primary production appears to be limited to sites where N effects are known to be negative.

p. 31, paragraph 1: Most ecosystems in the US are sensitive to N deposition in the sense that growth increases might occur. In some cases this will be “bad”, as in exotic species invasions, and in some cases this will be “good” as in commercial forests.

p. 32, GIS mapping: I firmly believe that the GIS mapping exercise should also include a thorough analysis of potential effects of increased N deposition on C sequestration.

p. 36, Table 3-5: Indicators should include foliar N concentration and should be stratified by species. We have reasonably good data on foliar N levels for deficiency, and even toxicity levels for many species. Foliar N should be included in the indicator list.

p. 38: The list of prospective sites here should include fast growing forests of the southeast (loblolly pine, for example) where N inputs may well cause greater growth and C sequestration. Also should include the San Bernardino sites in southern California where N saturation is extreme. San Bernardino’s are buried in Table 3-7, but should be included here. This list is biased toward sites with negative effects and incomplete.

p. 44: Lake Tahoe should include forest effects, were forests are thought to be typically N limited. Lake Tahoe is also somewhat unique in that much of the pollution enters the lake directly since ratio of land to water in the watershed is very low.

#### References:

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

Jenkinson, D.W., K. Goulding, and D.S. Powlson. 1999. Nitrogen deposition and carbon sequestration. *Nature*

Nadelhoffer, K.J., B.A. Emmett, P. Gunderson, O.J. Kjønaas, C.J. Koopmans, P. Schleppl, A. Tietema and R.F. Wright. 1999. Nitrogen deposition makes a minor contribution to carbon sequestration in temperate forests. *Nature* 398: 145-148.

Pregitzer, K.S., A.J. Burton, D.R. Zak, and A.F. Talhelm. 2009. Simulated chronic nitrogen deposition increases carbon storage in Northern Temperate forests. *Global Change Biology* 14: 142-153.

## **Dr. Donna Kenski**

I had 2 assignments for this document so I'll address those first:

I was asked to comment specifically on Sec. 2.3.1. While it minimally fulfilled its stated purpose (i.e., to provide an overview of the risk assessment framework), it was too abbreviated (4 paragraphs!) to be as effective as it could be. Section 3.7 actually did a much more comprehensive and coherent job explaining the risk assessment and its application to the standard-setting process. In fact, Section 3.7 was the first time I felt like this document actually pulled all the pieces together and made a case for how the process should work. Consequently I recommend beefing up Sec. 2.3.1 with much of the text from Sec. 3.7 (the bulleted lists especially, as well as the revised version of Fig. 2.1).

My second assignment was Charge question 11: "Additional ecological/welfare effects due to NO<sub>x</sub> and Sox emissions that we do not currently anticipate evaluating in detail in this review include the following:

Nitrogen saturation,

Maple decline,

Ammonia air deposition and toxicity to native mussels,

Relationships between acidity/nutrient enrichment and mercury methylation,

Sensitive areas for acidity/nutrient enrichment impacts, identified from biogeochemical characteristics, and

Climate change effects due to N<sub>2</sub>O.

Does the Panel agree that these represent lower priority effects for the current assessment? If not, what does the Panel recommend?"

While I agree that these MAY be lower priority effects, I don't think they necessarily should be neglected altogether. Although the plan says that N<sub>2</sub>O, for example, will be included in the scope of the review in a non-quantitative way (p. 13, Sec. 2.3.2), it never mentions how these lower priority effects will be included, even in a qualitative way. The rest of the plan seems to focus solely on the modeling of the 4 defined risks (p. 15, aquatic and terrestrial acidification and nitrogen enrichment) and never circles back to pick up these other effects. At what point do they get incorporated and how? If this is part of step 5, it was not obvious. It should be explicitly stated.

Of these lower priority effects, the only one that seemed really tangential is the ammonia deposition and toxicity to mussels. It is well established that mussels are sensitive to ammonia in water, but I was unable to find any papers on ammonia deposition and subsequent impact on surface water concentrations. These should be cited, at least in Appendix A where the subject is briefly reviewed.

Other general comments:

I found this document very difficult to follow, but I'm not sure exactly why. Organizationally it seemed sound. There were some terminology issues that complicated the presentation. For example, total reactive nitrogen is defined in the glossary as oxidized and reduced species as well as organic compounds, but this is not the standard terminology in atmospheric chemistry, I believe (it may be in the deposition/ecological world). Seinfeld and Pandis (in *Atmospheric Chemistry and Physics*) and many others define reactive nitrogen as NO<sub>y</sub> (oxidized and organic species), not including reduced nitrogen species. It seemed like the document was inconsistent, using reactive nitrogen to mean NO<sub>y</sub> sometimes and other times to mean NO<sub>y</sub> plus NH<sub>x</sub>. For example, p. 11, Sec. 2.3.2, 4<sup>th</sup> line, defines total reactive nitrogen as NO<sub>y</sub> (or at least I think that's what it's trying to do, with an unfortunate typo using reduced for reactive). Then again on page 23, end of 1<sup>st</sup> paragraph, it says EPA plans to evaluate the contribution of NO<sub>x</sub> to total reactive nitrogen relative to reduced forms of nitrogen. So I'm left quite confused about how and when ammonia/um will or won't be incorporated into this assessment. This leads to a larger concern when we get to the very end (p. 59), where some useful bullets itemize questions about the levels and forms of the standard that might be considered. Somewhat ominously, it sounds as if these questions might not be discussed as part of the risk assessment but only as part of the policy assessment (i.e., the ANPR). I hope that is not the case and that the coming risk/exposure assessment will discuss these options thoroughly. More important, it is not clear from this short list whether the indicator itself will be evaluated, and again whether it will include total nitrogen effects, versus total reactive nitrogen (NO<sub>y</sub> + NH<sub>x</sub> or just NO<sub>y</sub>??).

The general approach, especially the reliance on GIS mapping, seems sound, but the real question is how much data exist to support this effort in a meaningful way. Not possible to assess this from the Scope and Methods Plan.

Appendix B is too short to be useful. Rather than just describing the PnET-N-DNDC model and its input parameters, it should have incorporated model results.

## Dr. Naresh Kumar

*Charge Question 1. In outlining the scope of this risk/exposure assessment, we have created a flow diagram that represents how nitrogen and sulfur compounds move from ‘source to dose’ in the environment (see Figure 2-1). How adequately does this conceptual model for evaluating risks due to deposition-related ecological effects characterize what should be covered in the scope for this assessment?*

The conceptual model shown in Figure 2-1 is a simple, but efficient way to represent risk assessment framework for deposition-related ecological effects. Various endpoints, such as “acidification”, “nutrient enrichment”, “climate change”, etc. are listed outside the dotted line suggesting that these will be described in detail in the assessment document. However, later it is mentioned that the N<sub>2</sub>O effect (climate change) is outside the scope of this assessment and will be included only quantitatively. It is fine to do so, but Figure 2-1 should be modified to make it clear which endpoints will be assessed only quantitatively (one suggestion is to italicize such endpoints).

In Section 2.3, Page 8, where the N<sub>2</sub>O effect is discussed the first time in the text, a discussion on the scope of assessment for N<sub>2</sub>O effect should be included. For example, it should be made clear whether the N<sub>2</sub>O effect will be part of final deliberations for review/revision of the NAAQS for NO<sub>x</sub> and SO<sub>x</sub>.

Also, carbon sequestration should be added as an endpoint under “Nutrient Enrichment” in Figure 2-1 and a discussion should be included in the text describing this effect. Similar to the N<sub>2</sub>O effect, it should be made clear whether the carbon sequestration effect will be part of final deliberations for review/revision of the NAAQS for NO<sub>x</sub> and SO<sub>x</sub>.

## Dr. Myron J. Mitchell

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

### General Comments

More consistency is needed throughout the document on the different definitions and forms of chemical species including the grouping of these chemical species into classes. The linkage of acidification with cation (e.g., Ca and Mg) nutrient depletion and the mobilization of toxic cations (e.g., aluminum) needs to occur earlier in the document so that the total ramification of acidification is clearer. The document needs considerable work with respect to consistency and the use of precise descriptions. Some statements lack clarity. The criteria for selecting case studies are discussed, but an unambiguous delineation of the importance of specific criteria in making these selections is needed. Throughout the document there are statements about the needs for data. It would be better to state that “information” is what is required. Data availability without previous interpretation and analyses are less useful than those data sets that have been evaluated. Those data sets that have been evaluated by multiple approaches would be especially useful. Some reference to the European experiences in developing methods for Risk/Exposure Assessment might be insightful including the relatively wide application of the MAGIC model at various European sites.

Within these proposed scope and methods in some instances the plan seems very ambitious and I am not certain that completion is possible within the time frame of the assessment and with the resources available to do these tasks. This issue may be especially important in evaluating impacts on ecosystem services. The document needs to more clearly articulate priorities with respect to various approaches. I would suggest that it may be most facile to focus especially on the case studies to show most clearly the linkages between sulfur and nitrogen deposition and adverse damage to ecosystems.

### Specific Comments

| Page | Comment                                                                                                                                                                                                                                                                                                                                                                                             |
|------|-----------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------------|
| 2    | It is not clear whether N <sub>2</sub> O is included with the definitions of “oxides of nitrogen” and or “nitrogen oxides.” On page 11 it is stated that “the family of nitrogen oxides includes any gaseous combination of nitrogen and oxygen, e.g., NO <sub>2</sub> , NO, N <sub>2</sub> O, N <sub>2</sub> O <sub>3</sub> , N <sub>2</sub> O <sub>4</sub> , and N <sub>2</sub> O <sub>5</sub> .” |
| 6    | Change to:    in previous NAAQS <u>reviews on nitrogen or sulfur</u>                                                                                                                                                                                                                                                                                                                                |

- 11 Change to: In an analytical approach unconstrained by data and other resource limitations, one could envision a comprehensive risk assessment covering all potentially affected ecosystems and all scientifically supported effects on those ecosystems in the United States.
- 11 The identification of the N chemical species provided below needs to be better aligned with the definitions of N chemical species given on page 2: “The sum of mono-nitrogen oxides, NO<sub>2</sub> and NO, typically are referred to as nitrogen oxides (NO<sub>x</sub>) in the atmospheric science community. More formally, the family of nitrogen oxides includes any gaseous combination of nitrogen and oxygen, e.g., NO<sub>2</sub>, NO, N<sub>2</sub>O, N<sub>2</sub>O<sub>3</sub>, N<sub>2</sub>O<sub>4</sub>, and N<sub>2</sub>O<sub>5</sub>. Total reduced nitrogen (NO<sub>y</sub>) includes all nitrogen oxides as well as gaseous and particulate nitrate species such as HNO<sub>3</sub>, PAN, and aerosol phase ammonium nitrates. Reduced atmospheric nitrogen species include ammonia gas (NH<sub>3</sub>) and ammonium ion (NH<sub>4</sub><sup>+</sup>), the sum of which is referred to as NH<sub>x</sub>.”
- 13 The following statement is confusing with respect to how N<sub>2</sub>O is treated in this assessment: “Since the definition of “welfare effects” includes effects on climate [CAA Section 302(h)], we will include N<sub>2</sub>O within the scope of this review. However, it is most appropriate to analyze the role of N<sub>2</sub>O in anthropogenic climate change in the context of all of the greenhouse gases. Since that is outside the scope of this review, it will not be a quantitative part of this assessment.”
- 13 I don’t believe that generally the SO<sub>2</sub> or SO<sub>x</sub> includes particulate sulfate particles as stated: “SO<sub>2</sub> is one of a group of substances known as SO<sub>x</sub>, which include multiple gaseous (e.g., SO<sub>2</sub>, SO, SO<sub>3</sub>, S<sub>2</sub>O<sub>3</sub>, S<sub>2</sub>O<sub>7</sub>) and particulate (e.g., ammonium sulfate) species (**Figure 2-4**).”
- 21 Is this statement really necessary: “In addition to these four effects, we plan to address, as appropriate and within our time constraints, impacts associated with nitrous oxide (N<sub>2</sub>O) and the influence of sulfur enrichment on methylmercury production.”
- 22 This statement needs to be reworded to clearly suggest the challenge associated with making an assessment of impacts on a diverse set of ecosystem types over a range of atmospheric inputs: “The anticipated spatial extent and diversity of ecological effects due to deposition of nitrogen and sulfur do not facilitate a nationwide analysis.”
- 24 Change to: case study areas, data may not be sufficient to perform a quantitative assessment for each
- 28 In Table 3-2, why are K and Na not included with Ca and Mg as base cations?
- 28 Change to: “exchange capacity (CEC) are more widely available at the present time than information on biological indicators”

- 28 The following statement implies that CASTNET underestimates dry deposition of nitrogen. If this is the case, further explanation is warranted: “This creates complications in developing estimates for total nitrogen deposition levels because dry-deposition data sources will likely be underestimated.”
- 29 The following statement is not true since there is a linkage with Ca availability and tree species (e.g., sugar maple) that produce litter this has higher N mineralization and nitrification rates: “For terrestrial ecosystems, low calcium to nitrogen ratios in soils are commonly related to increased nitrification and potential increases in soil acidity and releases in NO<sub>3</sub> to receiving waters; however, these measurements are not always widely available.” Literature examples include:
- Christopher, S.F., B.D. Page, J.L. Campbell and M.J. Mitchell. 2006. Contrasting stream water NO<sub>3</sub><sup>-</sup> and Ca<sup>2+</sup> in two nearly adjacent catchments: the role of soil Ca and forest vegetation. *Global Change Biology* 12:364-381.
- Page, B.D. and M.J. Mitchell. 2008. Influences of a calcium gradient on soil inorganic nitrogen in the Adirondack Mountains, New York. *Ecological Applications*. (In Press)
- 29 Change to: For aquatic ecosystems, the indicators for “nutrient enrichment” effects reflect a combination of inputs from various sources
- 29 Not sure what “are encouraged” implies in the following: “are encouraged for inclusion in numeric criteria as part of EPA-approved state water quality standards (U.S. EPA, 2000)”.
- 29 Clarification is needed on the “data-fusion approach.”
- 29 In table 3-3 change to: “Reflects a combination of inputs from various sources (air, discharges to water, diffuse runoff, and groundwater inputs)”.
- 29 In table 3-3, the following statement (Repeated twice) is confusing: “Relative role of air deposition should ideally be compared with air deposition data and also with available (preferably multi-media) models.”
- How can the models be separated from air deposition data since the models are needed to calculate deposition?
- 30 Is it correct to include within ecosystem services “cultural services including spiritual or religious values, aesthetic values?”
- 30 What is meant by “type of environmental system?”
- 30 It is not clear how the following approach will produce the needed outcome:  
**“Identify Databases of Indicator Conditions:** The indicators selected will relate to

available compendiums of literature abstracts or actual database systems (as stand alone files or accessed through Web portals) to provide readily available and transparent ways to document the nature of the indicators and the indicator conditions used to define the environmental impairments.”

“Compendiums” should be “compendia.”

- 30 Change to: **Identify and Address Temporal Issues:** Different ecoregions, landscapes, biological provinces will have differing degrees of susceptibility to impairments or differing recovery potential, depending on edaphic characteristics, past land use and/or pollution histories.
- 30 For the following statement, be more explicit on the actual times needed for recovery (fairly rapid and much longer recovery times need some approximate values):  
“Some ecological systems may be capable of fairly rapid recovery responses once pollutant loadings are significantly abated; other systems, such as larger estuarine aquatic systems, may require much longer recovery times.”
- 31 Provide better description versus the following jargon: or at least define these terms “potential near-field and far-field linkages.”
- 31 The terms in Figure 3-2 should be reflected in the text.
- 31 Change to: Inland acid-sensitive waters in the eastern United States and nitrogen-sensitive ecosystems in the Rocky Mountains and other parts of the western United States may require large-scale, special-area assessments.
- 31 Clarify why this area is special (Is this different from the sensitivity of other areas?):  
“Ecosystem effects in special areas”
- 31 How could linkages not be geographically significant? Note the statement: “If the linkages are geographically significant”
- 32 Why this specific example: (e.g., local research of MeHg formation in Devil’s Lake, WI).  
  
If this example is used, at least a citation is needed.
- 32 Change to: Of special interest will be the characterization of linkages that can be used for synthesizing results for the entire U.S.
- 32 Why “may” in the following statement: “Information that deals with special case study areas (e.g., the Adirondack Mountains or special alpine and sub-alpine”  
  
Previous statements have suggested the importance of focusing on sensitive systems.
- 32 For heading “3.2.2 GIS Mapping” delete mapping; this is redundant.

- 32           Reword for clarity and precision: “To describe the national picture, we plan to map the locations of those sensitive ecosystems identified in Section 3.2.1 and identify the characteristics of the biological and biogeochemical properties that create the sensitivity.”
- 32           By “unmapped areas” does this imply areas in which an appropriate GIS is not available?
- 32           There are different issues associated with scale and defining the boundaries of a GIS. Clarify the following statement: “Sensitive areas can be identified at different spatial scales by using different approaches for defining the boundaries of the mapped units.”
- 32           The description under 3.2.2 needs considerable modification to clarify how GIS will be applied to this assessment.
- 33           The linkages need to be made more explicit in some components of: Figure 3-3. Documented biological, biogeochemical, and physiographic linkages.
- 34           Provide further details on the source of the “ESRI 8.3 data disks.”
- 35           It is stated that “Where case study or ecosystem-specific data are available, a subset of maps for the case study assessment area may be created.”
- Are only “data” needed rather than information associated with the interpretation of these data?
- 35           It is suggested that “Complementary to these efforts, we may use a statistical cluster analysis to group ecosystem units into similar sets. Clustering ecosystems might reduce the number of locations that need to be modeled to adequately characterize the variability in ecosystem response to changes in nitrogen and sulfur deposition.”
- It would also be helpful to identify key ecosystem attributes that are most critical in the classification and grouping of these ecosystems with respect to effects of N and S pollutants.
- 35           It is stated that “In selecting areas to assess ecological effects from air deposition, the SAB Ecological Effects Subcommittee (EES) suggests consideration of (1) clear quantifiable ecological effects due to air pollution, (2) the degree to which a significant component of ecological effects are attributable to air pollution, (3) the responsiveness of ecosystem services to changes in air pollution, (4) the cumulative impacts of multiple air pollutants, (5) the abundance of ecological effects and economic benefit cost analysis, and (6) the visibility to the public and value of resources at risk (U.S. EPA, 2005). While these recommendations were made in the context of a prospective cost-benefit analysis, many of these recommendations are sound in the context of our NAAQS risk analysis.”

Having some hierarchy in these selection criteria would be useful since there could be distinct differences among ecosystems on the relative value of each of these criteria.

- 35-37 In addition specific recommendations are provided from the ESS regarding selection criteria. These factors need to be blended into a more cohesive set of criteria for selection.
- 36-37 The information provided in Table 3-5 (Summary of Indicators, Mapping Layers, and Models for Targeted Ecosystems) is very diverse with respect to types of information, spatial scales, level of previous analyses, etc. It will be very useful to clearly indicate priorities in the selection of attributes from this table. This selection should be based upon well-identified criteria.
- 45 The following statement is weak: “Depending on the adequacy and abundance of data for areas, the evaluation may entail computer modeling, statistical analysis, or qualitative analysis.”
- Clearer criteria of the importance of the type and amount of information available need to be provided in selecting case study sites.
- 45 I am not sure that the following indicator is useful due the role of canopy exchange processes in markedly affecting the concentration and fluxes of N chemical species in throughfall: “Nitrate, ammonia, organic nitrogen throughfall deposition for terrestrial ecosystems.”
- 46 It is stated that: “We plan to evaluate the spatial adequacy of available monitoring data including GIS mapping of documented data to identify any meaningful spatial gaps.”
- What will be done if these spatial gaps are found?
- 46 It is stated that: “For each ecosystem effect, we plan to determine if there is a temporal dimension to exposure.”
- Isn’t there always a temporal dimension to exposure? Aren’t more critical issue aspects associated with non-linearities, thresholds, lag-times, etc.? Some of this is discussed in the following sentences.
- 46-47 Each of these sources of loading information has limitations. Including some details on the errors of these estimates would be helpful
- 47 In discussing the “CMAQ Deposition Modeling” the issues related to uncertainties associated with the calculation of deposition velocities as a function of surface types needs some mention with respect to the overall confidence in the predictions.

- 48 In discussing acidification the importance of the contribution of mobile anions (sulfate and nitrate) to the loss nutrient and toxic cations needs more emphasis. The pH effect is only a small part of the impacts.
- 50 An important criterion for selecting regions to model should include availability of previous modeling efforts.
- 50-51 Further clarity is needed in the section on “Assess Uncertainty in Loading and Exposure Computations.” For example does resolution include both temporal and spatial considerations? What is meant by “differing complexities?” Does this mean landscape complexities, complex processes, or something else? If more inventories of reactive N species are needed for the modeling, how will these inventories be done in the context of this effort?
- 51 Is it feasible to determine “the contributions from air deposition sources can be analyzed relative to all other major anthropogenic or natural sources for sensitive ecosystems across the country?”
- 51 How will the assessment balance the needs of doing rigorous analyses in one or possibly few regions with the greatest level of accuracy and precision versus a broader range of regions with lower accuracy and precision?
- 52 Be more specific in which parameters will be evaluated with respect to “response curves.”
- 52 More clearly differentiate how the Calculate Desired Exposure Endpoint compares with the determination of critical load including the criteria used in these determinations.
- 53 The following approach is quite vague and ambiguous: “The specific methods used to evaluate adversity will depend on the availability of data and methods for the indicators of interest related to acidification and nutrient enrichment and on an assessment of the appropriateness of each type of quantification in comparing different levels and forms of the standards. In our initial assessment of the available data, given the timeframe for this NAAQS review, we have determined that the most useful approaches will be those focusing on quantifying the link between changes in ecosystem indicators and ecosystem services.”
- Can some priorities be suggested on which indicators would have highest priority?
- 53 Doing the following is certainly a daunting challenge: “Thus, when ecosystem services are quantified and their ecological response functions to NO<sub>x</sub> and SO<sub>x</sub> are modeled, it is imperative that the entire bundle of services be evaluated, and that the linkages and tradeoffs among ecosystem services are included in the quantification (i.e., ecological tradeoff functions [ETFs]).”

- 54 Is it feasible to do the following within the time frame of this assessment?  
“Therefore, at this time, data mining will be central to developing at least a preliminary assessment of potential impacts of NO<sub>x</sub>/SO<sub>x</sub> deposition and acidity on ecosystem services. In the current plan, process-based models are being considered to be used to (1) synthesize/link the suite of ERFs and ETFs and (2) generate maps and summaries of ecosystem services and tradeoffs in response to current and future ambient air indicators for NO<sub>x</sub> and SO<sub>x</sub>. The collection of response and tradeoff functions will aid in the valuation of the services at risk to these criteria pollutants where possible.”
- 55 To accomplish the tasks that are stated might require a very selective choice of a region or a few regions where the available information is sufficient to do this work within a relatively short time:
1. Identify areas/regions of the country receiving high levels of NO<sub>x</sub>/SO<sub>x</sub> deposition and acidity impacts.
  2. In those regions, identify ecosystems sensitive to elevated levels of nitrogen and sulfur, using some common selection criteria.
  3. In those sensitive ecosystems, ask what ecosystem services are expected to be prevalent and “valued” (i.e., some subset of all potential services).
  4. In those areas, identify what data are available to develop ERFs and ETFs, at least to a qualitative degree that would enable production of spatial and temporal maps to identify different degrees of protection that would exist under alternative secondary NAAQS. Chan et al., (2006) produced such maps for several ecosystem services in the Central Coast ecoregion of California. The linkage and comparison of multiple ecosystem services in the region would provide information for consideration of tradeoff value of one service versus another.
- 58 The following evaluation approach is extremely ambitious:
- “1. Identify the relative contribution of loadings associated with atmospheric deposition of nitrogen and sulfur.
  2. Identify the most critical impacts from nitrogen and/or sulfur loadings (i.e., acidification, nutrient enrichment, or eutrophication).
  3. Identify the contribution to atmospheric loadings from total reactive nitrogen, NO<sub>x</sub>, and SO<sub>x</sub>.
  4. Identify the biogeochemical indicators/resources of concern in the assessment area and the ecosystem services associated with those indicators.
    - a. Determine the ecosystem service effects associated with the most-critical impacts.
    - b. Bundle ecosystem services to find common metrics for comparison across locations.
  5. Define the exposure-response (loading-response) functions (ERFs) for the ecological indicators of concern.

6. Estimate the loadings/exposures associated with current and alternative levels of the NO<sub>x</sub> and SO<sub>x</sub> standards (using CMAQ modeling).
  - a. Analyze the relationships between NO<sub>x</sub>, SO<sub>x</sub>, and other reactive forms of nitrogen.
  - b. Assess the impacts of meteorological variability on these relationships.
7. Estimate the ecosystem impacts associated with estimated loadings.
8. Convert estimates of individual ecosystem risks to common units using
  - a. economic valuation based on benefits transfer from existing literature estimates
  - b. biogeochemical equivalents using ecological tradeoff functions (ETFs).
9. Combine individual risk estimates to produce overall impact estimates”

Would it be more feasible to determine a hierarchal approach with respect to the selection of case study areas where the entire suite of approaches is utilized while for others a more limited assessment will be made?

59 The questions posed are valid and interesting, but addressing them will be a major task:

“How do alternative levels and forms of the standards relate to a given exposure metric?

What are the appropriate averaging times for alternative levels and forms of the standards?

What alternative levels of the standards should be considered?

Should there be alternative levels of the standards (i.e., individual NO<sub>x</sub> and SO<sub>x</sub> standards or a combined NO<sub>x</sub>/SO<sub>x</sub> standard)?

Do the ambient air indicator forms allow for site-specific protection while maintaining national consistency?

Does the ambient air indicator adequately account for the effects of total reactive nitrogen?

Does the form of the standard have an impact on the risk?

As I have said previously with respect to the overall approach, should some of these questions have greater priority?

A-1 In determining “Sensitive Ecosystems” having some criteria related to information availability would facilitate the assessment of “Targeted Ecosystem Effects.”

A-1 The susceptibility of the ecosystems to the depletion of nutrient cations should also be an important factor with respect to evaluating sensitivity.

A-1 In grouping ecosystems “statistical cluster analysis” should be supplemented with consideration of the major factors that are most critical in determining the impacts of N and S deposition. Can the cluster analysis include weighting of the most critical factors?

- A-2 In addressing the issues of N enrichment the importance of the generation of the mobile nitrate anion with respect to mobilization of toxic Al and H ions and loss of nutrient cations (especially Ca and Mg) needs to be included.
- A-2 The quantification and analyses of ecosystem services in the context of this assessment will be a major challenge.
- A-3  
to  
A-5 A major task will be the separation of atmospheric derived fixed N versus other N sources especially fertilizer and waste treatment in making evaluations for the larger coastal watersheds.
- A-5 The heading “A.1.3 Aquatic Sulfur Enrichment” is likely not appropriate since much of the discussion focuses on the generation of MeHg which most likely is predominantly generated in wetlands. Maybe a separate section on wetlands would be helpful.
- A-6 The following statement is not correct in the context of sulfate/MeHg relationships: “The “cause-and-effect relationship” between sulfur and mercury deposition from the atmosphere has been demonstrated in the lab and in small-scale field experiments”.
- A-6  
to  
A-7 The section on “Terrestrial Acidification Due to Nitrogen and Sulfur” with respect to the discussion on nutrient cation depletion should briefly indicate the importance of specific tree species such as sugar maple in this assessment.
- A-7 In discussing episodic acidification, some mention of climate linkages would be appropriate. This is relevant not only to snowmelt episodes but also the summer storm events, especially those following droughts.
- A-8 It would be better to indicate that this is “Sugar Maple Decline”. Some maple species are very resilient to atmospheric pollutants and nutrient depletion.
- A-9 Data suggests that sugar maple has higher Ca requirements than other dominant species in the northeastern forests of the U.S. making it more sensitive to Ca depletion. See for example: Page, B.D., T.D. Bullen and M.J. Mitchell. 2008. Influences of calcium availability and tree species on the cycling of Ca isotopes in soil, vegetation, and stream water. Biogeochemistry. DOI 10.1007/s10533-008-9188-

B-1

to

B-2

The importance of N<sub>2</sub>O emissions is due to its role as a green house gas. It may not be feasible to address fully the importance of this factor in this assessment without spending considerable effort integrating green house gas discussion into the document.

C-1

to

C-4

Some consideration is needed on the accuracy and precision of the estimates associated with the use of the CMAQ MODELING. How will the uncertainties associated with the modeling of the atmospheric deposition of sulfur and nitrogen affect the ability of the assessment to predict the effects of spatial and temporal patterns?

## Mr. Richard L. Poirot

### **Supplemental Post Meeting (4/2-3/08) Individual CASAC Comments on Secondary SO<sub>x</sub> & NO<sub>x</sub> Integrated Science Assessment and Ecological Risk/ Exposure Assessment Draft Scope and Methods Document**

Shortly after the 4/2-3/08 CASAC Review of the Secondary SO<sub>x</sub> & NO<sub>x</sub> Integrated Science Assessment and Ecological Risk/ Exposure Assessment Plan, it has come to my attention that EPA funding for several critical long-term atmospheric deposition-related monitoring and research programs, which have been invaluable to understanding the continuing effects of sulfur and nitrogen deposition, are scheduled to be substantially reduced or eliminated in the Administration's proposed 2009 budget. Specifically, the *Clean Air Status and Trends Network* (CASTNET) air quality and dry deposition network is scheduled for a 25% funding reduction in 2009, and the *Temporally-Integrated Monitoring of Ecosystems* and the *Long-Term Monitoring* (TIME/LTM) programs (which had recently been saved from proposed cuts in 2008, but) will be considered "completed" (i.e. terminated) in 2009. See attached letter from Assistant Administrator George Gray to Gary Lovett (chair of the Northeast Ecosystem Research Cooperative steering committee).

The timing for reduction and/or termination of these valuable long term programs is especially poor, since the Agency is currently considering secondary standards for SO<sub>x</sub> and NO<sub>x</sub>, which are being specifically focused on the environmental effects of S and N deposition. At the same time, large additional S and N emissions reductions are scheduled in the Eastern US under the Clean Air Interstate Rule (CAIR), while new sources of S and N emissions are coming on line in many western states, northern Mexico and western Canadian provinces.

The ISA and Ecological Risk/Exposure documents should be modified to reflect these planned changes in the Agency's sulfur and nitrogen deposition-related monitoring and research programs, clearly indicating:

- CASTNET, TIME & LTM are scheduled for large cuts or elimination in 2009,
- why the Agency plans to reduce or eliminate these long-term programs,
- how the Agency plans to evaluate environmental effects of future emission changes from CAIR, new SO<sub>x</sub> or NO<sub>x</sub> NAAQS, and/or other emissions changes,
- how the Agency plans to consider secondary SO<sub>x</sub>, NO<sub>x</sub> or PM standards in this or future NAAQS review cycles without the information on deposition and ecological effects that these programs provide.



UNITED STATES ENVIRONMENTAL PROTECTION AGENCY  
WASHINGTON, D.C. 20460

OFFICE OF  
RESEARCH AND DEVELOPMENT

Mr. Gary M. Lovett  
Chair, Steering Committee  
Northeastern Ecosystem Research Cooperative  
Cary Institute of Ecosystem Studies  
Box AB, Millbrook, NY 12545

Dear Mr. Lovett:

Thank you for your letter of January 29, 2008, requesting information on Environmental Protection Agency's (EPA) plans to fund the Temporally-Integrated Monitoring of Ecosystems and the Long-Term Monitoring (TIME/LTM) programs associated with EPA's Acid Rain Research Program. We are pleased to report that we are able to fund both of these programs in FY 2008 at the FY 2007 resource levels of seven hundred thousand dollars (\$720,000.00). As you suggest, the design of these monitoring programs is unique and the result of many years of research by the EPA's Office of Research and Development (ORD). These two programs have been extremely valuable in demonstrating the effects of the reduction of sulfur on aquatic resources, reflecting changes from the most recent Clean Air Act amendments.

It should be noted that the focus of the research in the TIME/LTM programs was on the design of the monitoring program, development of indicators to measure changes, and reporting on those changes as a means of verifying the intended results. With the FY 2008 funding, ORD will complete its defined goal for both of these programs. EPA will determine the appropriate disposition of these programs in FY 2009.

Again, thank you for your letter. If you have additional questions, please contact me or your staff may call James Blizzard, in EPA's Office of Congressional and Intergovernmental Relations, at 202-564-1695.

Best regards,

A handwritten signature in black ink that reads "George Gray".

Dr. George Gray  
Assistant Administrator  
Office of Research and Development

cc: Lek Kadeli  
Kevin Teichman  
Carl Mazza  
Becky Higgins

## Comments on SO<sub>x</sub>/NO<sub>x</sub> Eco-Risk Assessment Draft Scope & Methods Document

It was a pleasure to read through this planning document. It was clearly written and well-organized, founded on a sound conceptual model and guided by an excellent series of key policy-relevant questions (on pages 17 & 18). Because of the new and unfamiliar NAAQS review process, it's difficult to judge how far the information should be developed in each stage of the review (what belongs in the domains of the ISA, the risk assessment or the policy assessment) but it appears like the planned ecological risk/exposure assessment should provide an adequate basis for consideration of policy options for secondary NO<sub>x</sub> and SO<sub>x</sub> NAAQS.

At the same time, I think the potential for effective secondary standards could be substantially enhanced or limited by how broadly or narrowly the pollutant effects of interest are defined (for example including or excluding aerosol-phase effects), and by how much flexibility is allowed in considering the measured indicators by which compliance with the secondary standards will be determined (for example: SO<sub>2</sub>, total atmospheric sulfur, sulfur deposition, exceedance of critical loads for S or S+N deposition, etc.). Statements on page 56 and 57 of the plan imply that deposition-based indicators of ecological risk are not at all the same as and will need to be related to comparable ambient air concentration indicators - perhaps limited to the nominal gaseous criteria pollutants of sulfur dioxide and nitrogen dioxide. If that is the case, I doubt very much that any useful secondary standards can be developed for these pollutants, and I think this critically important issue and should be discussed in some detail at this review meeting.

Assuming this issue can be satisfactorily resolved, I also much admire the carefully reasoned conceptual model as well as the very ambitious approach for conducting the assessment. I don't personally have enough familiarity with the many types of atmospheric, geological, chemical, biological and socio-economic data and model parameters needed to conduct the proposed assessment, but it strikes me as a very challenging undertaking to first collect all these necessary layers of information and then and then make them all fit together to produce the proposed quantitative assessments, especially since each step in the process depends upon successful previous steps. I note the phrases "we plan to" or "we intend to" appear 43 times in the document, but there are no "we have done"s. In many cases, I assume you have already gathered some of these data and tested their use in some of the intended applications - i.e. have some sense that this approach will actually work. It would be helpful for reviewers to see some example intermediate demonstration products to give a better sense that this plan will actually work.

### Charge Questions 8-10:

*8. In the seven-step approach to the current conditions risk/exposure assessment, Step 5 (Section 3.5) discusses how to scale up case study areas to more spatially extensive sensitive areas, where appropriate. Does the Panel agree with this approach or can they suggest alternatives?*

It's not entirely clear how geographically limited your case study areas will be, but generally the process of scaling results to larger spatial domains should be reasonable if your case studies identify the key biogeochemical parameters associated with the effects, and if you have data with sufficient quality and resolution to identify similar conditions over larger regions. I imagine this

will often involve moving from intensive locations where detailed on-site measurements are available, to broader spatial areas where surrogate data estimates will need to be employed. Presumably it will be possible to test some of your projected scaled effects at other sites where detailed data are available. Possibly you could also test scalability by projecting site-specific results to higher or lower deposition rates from the recent past. It should also be possible and desirable to include some bounding estimates as you extrapolate results over space (& time).

*9. In the seven-step approach to the current conditions risk/exposure assessment, Step 6 (Section 3.6) outlines a path to assess the current conditions of sensitive ecosystems. How well does the Panel agree with the approach outlined for calculating response curves and utilizing mapping and ecosystem services to characterize current conditions or can the Panel recommend alternative approaches?*

*How well does the Panel agree with using ecosystem services to provide a common metric for comparing ecological risks due to nitrogen and sulfur deposition effects?*

*How well does the Panel agree with collecting current valuation studies to understand the value of bundled ecosystem services? Can the Panel recommend additional or alternative approaches?*

I have only general comments here, as I really don't know enough about the "ecosystem services" approach for assessing valuation. I think it should be possible to develop reasonable ecosystem response curves for a variety of endpoints as well as to provide a detailed descriptive summary of the various short and long-term consequences that are expected to result if critical loads or other such effects limits are exceeded. I would think this descriptive information of expected consequences could be carried through to future steps in the process, regardless of whether the "value of bundled ecosystem services" can be effectively communicated to your readers. I don't think it's absolutely necessary to directly compare/contrast the risks of S vs. N pollution (its not like we can have a standard for only one or the other), but if you have a way to present the very different effects of acidification & nitrification in common units than yes, this could be very helpful.

*10. In the seven-step approach to the current conditions risk/exposure assessment, Step 7 (Section 3.7) describes an approach to assess degrees of protection/levels of effects under alternative forms and levels of ambient NO<sub>x</sub> and SO<sub>x</sub> standards. This approach attempts to describe how the methods, models, and results of the current conditions risk/exposure assessment can inform our evaluation of the appropriate form(s) and level(s) of a national standard. How well does the Panel agree with the approach outlined in this section, the issues presented, and the 9 steps outlined to assess potential forms and levels of the standard? Please suggest any additional or alternative steps we should take into consideration.*

As indicated above, I have a major problem with the implied constraint on NO<sub>x</sub>/SO<sub>x</sub> indicators. If you are talking only SO<sub>2</sub> and NO<sub>2</sub> as NAAQS indicators, I think chances of useful secondary NAAQS are slim. A comparison of spatial & temporal fields of S & N emissions, ambient concentrations of gaseous SO<sub>2</sub>, NO, NO<sub>2</sub>, HNO<sub>3</sub>, NH<sub>3</sub>, aerosol SO<sub>4</sub>, NO<sub>3</sub>, NH<sub>4</sub> and wet & dry S and N (oxidized & reduced) deposition (recent past, current, and projected [post CAIR] future) should be a relatively straight-forward modeling exercise – which we would want to see confirmed to the extent possible with measurements. This kind of information can and should be conducted in parallel with and independently from the eco-risk assessment, and results

distributed as soon as feasible to this panel for review (if you can provide all of the above in gridded GIS data layers so much the better). At the same time, I think it would also be useful to consider the possible benefits of a wider range of indicators than SO<sub>2</sub> and NO<sub>2</sub>.

In making this broader assessment, I would encourage you to continually consider what the best ecological indicators would be and then consider whether we have in place adequate measurement programs to track them in the locations of concern. I would also encourage full consideration of the sources, effects and benefits of controlling reduced N, regardless of whether anticipated policy-relevant “NO<sub>x</sub>” NAAQS indicator metrics are likely to include it or not.

## Mr. David J. Shaw

### General Comments

#### *Outcome of REA*

Is this REA the same one that is described as due August 2008 (Table 2-1)? The REA Plan is difficult to assess in terms of what outcome to expect because it has so many uncertainties built-in, for instance “depending on available data”, or “if feasible” is used a lot. Also, who will do the actual assessment work, over what time period? What funds are available for the work? Who is the audience?

#### *Adopting a Longer View*

From what I have looked at so far, I think it is safe to say that most of the environmentally sensitive areas do not have adequate monitoring in place to track the changes that we will need to quantify when the next review of these pollutants become due in 5 -10 years. It seems appropriate to make it a requirement that this REA make specific recommendations on what needs to be in place to make the next assessment more adequately quantitative or populated with data. We don't want to see more sophisticated models with the same poor data. We do want to see a stronger data record of results on the ground.

This longer term view (5 to 10 year) is also an opportunity to examine in this REA what metrics would be more useful than simply going with those that are currently available, e.g. the base cation surplus is a better indicator than ANC or pH where DOC is changing like in the Adirondacks.

#### *Reliance on Modeling*

A model like CMAQ certainly can produce estimates of dry deposition, but one of the drawbacks is that there is no way to assess if these estimates are reasonable. For this reason, it probably makes more sense to emphasize those measurements and model predictions that we have the most confidence in, namely wet deposition. (Section 3.4.2, pages 46-47)

Using five years of model results certainly is better than relying on a single modeling year. Were these five years modeled in a consistent fashion – same version of MM5, consistent emission inventories, consistent CMAQ features/parameters, etc.? Have they been evaluated against all available ambient concentration and deposition measurements, both for individual years and for the entire five-year period? (Appendix C)

#### *Data Certainty*

Few, if any, of the references to existing data provide the years of data availability. The length and current-ness of records are critical to any assessment especially because the last decade has

been a time of significant emissions/deposition changes and ecosystem response signals have been detected.

Missing from the document is a discussion (with maps) of the documented areas of the US where sensitive ecosystems and high deposition rates of N and S have been identified (e.g. NAPAP Biennial Report to Congress: An Integrated Assessment, May 1998, Figure 19, page 55 ).

### ***PM & Ozone***

I feel it is important to address particulate matter and ozone in this ISA as well as the PM and ozone ISAs.

The omission of particulate phase  $\text{NO}_x$  and  $\text{SO}_x$  appears to limit 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.

Also, 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  $\text{NO}_x$  and  $\text{SO}_x$  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.

### ***Climate Change***

Climate change discussion appears to be limited to  $\text{N}_2\text{O}$ . I feel that the climate change issue may be addressed more appropriately by including foliar injury data (USDA).

Section 2.3.1, Figure 2-1, page 10: Consider amending this figure. Climate change may affect the “Characterization of Exposure” and “Characterization of Ecological Effects,” so consider putting arrows from “Climate Change” to these boxes. Suggest changing “Climate Change ( $\text{N}_2\text{O}$ )” slightly to “Climate Change ( $\text{N}_2\text{O}$ , etc.).”

### ***Charge Questions***

The 7 step approach seems thorough, but I do not feel that the gaps and uncertainties are properly described.

Identifying documented effects seems appropriate, but I feel that not all indicators are identified. Specifically, foliar injury (USDA) and base cation surplus.

### **Specific Comments**

Section 2.3, page 8: Why is the document disregarding damage to materials, including decay of buildings, statues, and sculptures that are part of our national heritage this time around?

Section 2.3.2, pages 11-13: Although N<sub>2</sub>O is included in this review process, it is not part of NO<sub>y</sub>. Nor does it play a role in N deposition, so while it could be included elsewhere in this document it does not really belong in this section.

Section 2.4, last bullet on page 17: Is it necessary to be able to distinguish NO<sub>x</sub> from total reactive N? Isn't the purpose of this to examine all oxides of N?

Section 3.1.1, page 28: add 'base cation surplus' to Table 3-2. Key indicators of nutrient enrichment. This first paragraph is about deposition and applies generically to both acidification and nutrient enrichment issues.

Section 3.3, page 36-37: Table 3-5 More detail needed on the indicator data. Add dates of the mapping layers, whether point or polygon data, the number of data points or polygons in the study area (maybe the whole US), and the resolution of the base data.

Section 3.3, page 38: I feel that including the Catskills of New York will provide important information and data.

Section 3.4.1, page 45: Selection of ANC as an indicator. Have all sensitive regions and case study areas been evaluated independently for selection of best indicators? Just because ANC is most widely available, it may not necessarily be the best, e.g. see discussion on using base cation surplus for Adirondack streams in the ISA. Also the three first bullets of indicators are not really indicators (see page A-4 paragraph for indicators).

Section 3.4.2, page 47: The maps are too small.

There are many acronyms throughout this document, and not all of them are commonly used. Please include a listing of these.

## Dr. Kathleen Weathers

This is a useful document for catalyzing discussion and getting feedback from the committee. The approach relies almost exclusively on process-based and spatial modeling analysis. A short discussion of what—if any—other types of analytical and assessment tools are available would be useful.

Specific and general comments, questions and suggestions are listed below.

### Section 2.3.1

I applaud the use of a conceptual model and framework.

Explicitly missing from the conceptual framework, however, is the key link (flow arrow) between concentrations and deposition. Also, the significance of the arrows and boxes should be identified (e.g., what's the distinction between the dotted and solid lines surrounding boxes. And the arrows?

What's the difference between ecosystem effects and ecosystem responses, or services (in boxes), for that matter?

I think that it would be helpful to define general ecosystem functions (e.g., productivity, nutrient cycling, etc.) and then point to places where the deposition of S and N has been demonstrated to alter them.

Page 1: Consider altering the sentence beginning with “Our plan...” to: “...and the levels of deposition,” *and the environmental effects on ecosystems of this deposited material.*”

Page 1: “Along with these case studies, we plan to conduct statistical and spatial characterizations...” Are these analyses intended to be new work/research?

Section 2.1: The legislative mandate reads to me as the request for determination of critical loads, albeit within a narrow range.

Section 2.2, page 6 awkward phrase: ....importance of acid neutralizing capacity in surface water acidification.

Page 8, 2<sup>nd</sup> para: *tropospheric ozone*

Page 8, last para: “Against this broad background...” awkward and, in places incorrect.

Page 9, “In this current review...appropriateness of NO<sub>2</sub> as an indicator...” I'm not sure what this means.

Words such as harmful (page 11) should be modified by ecological or environmental.

The consideration of critical load analysis as an ecosystem-type specific risk may be more useful than considering it as a site-specific risk.

Page 11, the paragraph beginning with “Each component of the framework...” isn’t clear.

Fig 2-2 might note explicitly the formation of ammonium sulfate, since both N and S are of interest here.

2.3.3. The first couple of sentences are awkward. Also, acidification of the environment should include lowering the natural pH of rain, water bodies and/or terrestrial ecosystems.

2.3.4: are the deposition of NO<sub>x</sub> and SO<sub>x</sub> meant to infer the deposition of nitric and sulfuric acids? If not the second sentence is not right.

2.3.4: Be specific, for example: “Nitrogen and sulfur enrichment represents a continuum of effects...and it can be characterized as positive (increase in environmental parameter y with enrichment, or additional N or S, or with incremental additions of N or S) or negative (decrease in environmental parameter y with increase in N or S, etc)

Say clearly why methylmercury is being considered here vs other linked biogeochemical interactions (the effect of sulfur on phosphorous release/availability in aquatic systems, for example).

Figure 2.5: consider depicting feedbacks and linked processes

#### **Section 2.4:**

Clarify...”rather than on the effects of aerosol NO<sub>x</sub> and SO<sub>x</sub> that remain in the atmosphere.”

Page 17, second bullet: responses = effects? and variability is meant to be spatial and temporal, correct?

Page 18, second set of bullets:

Identifying important chemical species in the atmosphere? Important means relative to other species, because they can cause environmental effects? Over space and time?

Page 18, last bullet: Changes in land use might be added to the meteorologic and climate considerations

#### **Section 3: Seven-step approach**

The 7-step approach is appealing. It uses both spatial and mechanistic models to explore inputs of N and S across the US as well as the effects of N and S deposition. I remain concerned, however, that (1) the data that are necessary for many of the analyses do not exist or (2) available data are not at an appropriate spatial or temporal scale and/or cannot readily or reasonably be

linked together or extrapolated using geostatistical or other tools. (3) Finally, I cannot tell whether or how experimental data, for example N or S or combined N and S additions would be used to inform this analysis.

Since both the use of biogeochemical process models in new geographic locations as well as the creation of GIS-based models and analysis are active areas of scientific research, to what extent is it necessary that these model analyses and extrapolations appear in peer reviewed publication before they can be effectively used in this assessment? In addition, the steps that will be used to validate or verify model results should be identified.

Under *Key Indicators of Nutrient Enrichment*: What's the basis of the suggestion that wet-deposition monitoring stations can provide more... an extensive range of nitrogen species than is possible for dry-deposition monitoring stations?

As mentioned in my review of the science assessment, throughfall N may not reflect N deposition levels. Also, C:N in soils have been related to nitrate in surface waters draining watersheds.

Table 3-3: Technically, there aren't "dry deposition monitoring" networks, rather there exist air concentration monitoring networks and those data are used to estimate dry deposition.

See comment above about indicators and throughfall. Sulfur has been used successfully as an index of wet+dry deposition in the eastern US.

Recovery should be defined clearly and carefully for the purposes of this assessment.

What's the operative definition of a multi-media model?

#### **Step 5 (Section 3.5):**

The spatial extrapolation of case study areas is an interesting approach; my answer to whether I agree with the approach depends upon answers to several questions. It is not clear to me the basis upon which case study areas could be scaled, or whether the interpolation techniques alluded to are appropriate spatial extrapolation tools. Presumably deposition output from the CMAQ model will be mapped on to the sensitive areas, for instance. Are the spatial scales comparable?

#### **Step 6 (Section 3.6):**

The goal of creating response curves needs more explanation. Is the goal to identify thresholds, or identify inflection points? What's a desired exposure endpoint? How will, or how could, published loading experiments be used to help identify response curves?

An example of "quantifying the link between changes in ecosystem indicators and ecosystem services" would make easier a discussion on the utility of this approach.

I concur with the idea of using the Millennium Ecosystem Assessment's definition of ecosystem services.

I think that parts of the mapping steps identified on page 55 are a good start:

It makes sense to me that the first data layer should be a total (wet + dry) S and N deposition map, apparently generated from CMAQ.

The acidity impact datalayer is presumably derived, but from which data or datalayers? And, from here on out with the mapping exercises, the devil is in the details. For example, what are the common selection criteria? How will the prevalent ecosystem services be identified? A highlighted example from the Chan et al. 2006 paper would aid this discussion.

The valuation of ecosystem services is out of the realm of my direct expertise, but it seems to me that there is always the danger of undervaluing ecosystem services, especially when considering biogeochemical cycling of multiple, interacting elements.

### **Step 7 (Section 3.7):**

Based on the heterogeneity in S and N loading, ecosystem sensitivity and responses across the United States, I think it very important to include as part of the secondary NAAQS a "form that allows for consideration of regional heterogeneity."

Resilience (and recovery) will need to be defined.

The need to relate ambient concentrations with actual deposition loads appears again in this section.

In the list of associated issues, "If total nitrogen..." concentration, content in what?

I agree that it will be most fruitful to do intensive analyses on smaller regions.

Will the proposed analyses be done on watersheds? Or on ecosystems with some other boundary and if so, what and why? It would be useful to define boundaries for these assessment areas.

Many, if not all, of the 9 steps entail significant analysis (which is evident to those who will be working on this document, of course). Are all steps equally important to come to policy relevant conclusions? Is it possible to identify what the most "sensitive" steps are, meaning which ones have the greatest influence on the policy-relevant conclusions. What is the relative importance of assessing the impacts of meteorological variability on loadings and exposures across the US (presumably) vs analyzing the relationships between NO<sub>x</sub>, SO<sub>x</sub> and other reactive forms of N (in the atmosphere)? My assessment of the approach depends, in part, on answers to these questions.

# **Attachment I: Comments on First External Review Draft of “Integrated Science Assessment for Oxides of Nitrogen and Sulfur: Environmental Criteria”**

**Prepared for the  
Alliance of Automobile Manufacturers**

**by**

**Jon M. Heuss**

**Air Improvement Resource, Inc.**

**March 17, 2008**

## **Executive Summary**

The effects of acidification and nitrogen-nutrient deposition on ecosystems are the primary focus of the draft Integrated Science Assessment (ISA). Although the U. S. Environmental Protection Agency (EPA) is combining the review of oxides of nitrogen (NO<sub>x</sub>) and sulfur (SO<sub>x</sub>) welfare effects in this one document, these Air Improvement Resource, Inc. (AIR) comments concentrate on the science that will influence the NO<sub>x</sub> secondary standard decision. There was substantial evidence of NO<sub>x</sub>-related acidification of freshwater streams and lakes and eutrophication of estuaries and coastal waters in the last review completed in 1996. However, at that time, the Administrator concluded that adoption of a nationally-uniform secondary standard would not be an effective way of approaching these problems.

In the last decade there has been substantial new information on acidification effects on terrestrial and aquatic ecosystems and eutrophication effects of excess nitrogen. There have also been major reductions in man-made NO<sub>x</sub> and SO<sub>x</sub> emissions as well as control programs put in place for additional NO<sub>x</sub> and SO<sub>x</sub> reductions in the coming decades.

These AIR, Inc. comments on the first draft ISA and its 10 Annexes focus on the science and data most critical to the policy decisions that will ensue once the ISA is completed. The most important changes needed to the draft ISA are:

- An explicit listing and discussion of potential indicators needs to be included in the ISA. The science that informs the important threshold question in the review - what is the appropriate indicator or indicators? – needs to be fully vetted in the ISA.
- There are both technical and legal considerations in choosing an appropriate indicator; the ISA needs to include a short discussion of the legal questions since the Clean Air Act presupposes nationally uniform primary and secondary national ambient air quality standards (NAAQS).
- The ISA should evaluate both reduced and oxidized forms of reactive nitrogen equally. The discussion needs to start with a description and understanding of the natural N cycle.

- The beneficial effects of anthropogenic nitrogen deposition as it relates to fertilization/carbon sequestration/climate change needs to be evaluated in the ISA so it can be weighed against the potential adverse effects in the policy considerations.
- Similarities and differences between sulfate and nitrate behavior and effects need to be clearly delineated.
- The discussion of NO<sub>x</sub> emissions from soil and lightning is missing important papers that demonstrate these sources result in significantly higher emissions than noted in the ISA
- Differences in the N deposition footprint between ground-level and elevated sources need to be discussed.
- The discussion of direct deposition to vegetation should include the fact that current levels of deposition directly to vegetation are not likely to cause risk or harm.
- If deposition-based secondary standards are to be considered, a discussion of the quality and completeness of deposition measurements should be included in the ISA.
- The discussion of atmospheric modeling is too general; the focus should be on the performance of the CMAQ model (or other models) that will be used in the risk assessment.
- Issues with the critical loads approach need to be explicitly discussed in the body of the ISA. For example, although the Europeans have been developing critical loads as a tool for evaluating adversity and setting policy, they do it in a cost-benefit framework. This is not consistent with the U. S. NAAQS paradigm.
- The implications of the fact that many sources contribute to eutrophication need to be discussed. .
  - Eutrophication is not primarily an air pollution problem; it involves not only combustion sources of NO<sub>x</sub>, but also agricultural practices, fertilizer use, treatment of human waste, and treatment of animal waste.
  - The wide range of atmospheric contributions, sensitivities of different water resources, and contributions from non-atmospheric sources makes using a national ambient air quality standard to address eutrophication problematic.

Based on the comments below, the draft ISA needs extensive revision before it can serve as the basis for policy decisions regarding secondary NAAQS for NO<sub>x</sub> and SO<sub>x</sub>.

## Introduction

The draft “Integrated Science Assessment for Oxides of Nitrogen and Sulfur: Environmental Criteria”<sup>1</sup> (ISA) is designed to be a concise synthesis and evaluation of the most policy-relevant science for use in EPA’s decision-making process concerning appropriate secondary air quality standards for oxides of nitrogen (NO<sub>x</sub>) and sulfur (SO<sub>x</sub>). Since NO<sub>x</sub> and SO<sub>x</sub> emissions contribute to ambient particulate matter (PM), EPA will be considering some NO<sub>x</sub> and SO<sub>x</sub>-related welfare effects such as visibility reduction in the review of the secondary PM standards. The effects of acidification and nitrogen-nutrient deposition on ecosystems are the primary focus of this draft ISA. Although EPA is combining the review of NO<sub>x</sub> and SO<sub>x</sub> effects in this one document, these AIR, Inc. comments concentrate on the science that will influence the NO<sub>x</sub> secondary standard decision.

In October 1996, in the previous review of the NO<sub>x</sub> secondary standard, EPA decided to set the secondary standard identical to the primary standard. Although there was substantial evidence of NO<sub>x</sub>-related acidification of freshwater streams and lakes and eutrophication of estuaries and coastal waters, the Administrator concluded that adoption of a nationally-uniform secondary standard would not be an effective way of approaching these problems.<sup>2</sup> The Administrator indicated that the decision would be re-visited in the next review and that, meanwhile, the Title IV program that addresses acid deposition and other national and regional programs will be reducing NO<sub>x</sub> emissions. At that time, consideration was given to an acid deposition standard, but CASAC<sup>3</sup> and EPA staff<sup>4</sup> agreed that the available scientific information did not provide an adequate basis for standard setting purposes.

In 1995, an EPA report to Congress<sup>5</sup> on the feasibility of acid deposition standards had indicated that establishing acid deposition standards for nitrogen and sulfur deposition may at some point in the future be technically feasible. In addition, a 2004 National Research Council report<sup>6</sup> had concluded that concentration-based standards are inappropriate for some resources that are at risk from air pollutants, including soils, groundwater, surface waters, and coastal ecosystems. The NRC report indicated that deposition standards would be more appropriate for such resources and noted the “critical loads” concept, in particular, as one candidate approach.

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<sup>1</sup> U. S. Environmental Protection Agency, First External Review Draft of “Integrated Science Assessment for Oxides of Nitrogen and Sulfur: Environmental Criteria,” EPA/600/R-07/145A, December 2007; U. S. Environmental Protection Agency, First External Review Draft of “Annexes for the Integrated Science Assessment for Oxides of Nitrogen and Sulfur: Environmental Criteria,” EPA/600/R-07/145bA, January 2008.

<sup>2</sup> 61 Federal Register 52852, October 8, 1996.

<sup>3</sup> G. T. Wolff, CASAC closure letter for the 1995 OAQPS Staff Paper, addressed to U. S. EPA Administrator Carol Browner, August 22, 1995.

<sup>4</sup> U. S. Environmental protection Agency, Review of the National Ambient Air Quality Standards for Nitrogen Dioxide: Assessment of Scientific and Technical Information, EPA-452/R-95-005, September 1995.

<sup>5</sup> U. S. Environmental Protection Agency, Acid Deposition Feasibility Study Report to Congress, 1995

<sup>6</sup> National Research Council, Air Quality Management in the United States, National Academy Press, Washington DC, 2004.

In the decade and more since the last NO<sub>x</sub> Criteria Document (1993),<sup>7</sup> there has been substantial new information on acidification effects on terrestrial and aquatic systems and eutrophication effects of excess nitrogen. There have also been major reductions in man-made NO<sub>x</sub> and SO<sub>x</sub> emissions as well as control programs put in place for additional NO<sub>x</sub> and SO<sub>x</sub> reductions in the coming decades.

Given this background, the AIR, Inc. comments on the ISA and its 10 Annexes focus on the science and data most critical to the policy decisions that will ensue once the ISA is completed. The following provides both important general comments and specific comments on the draft ISA.

## General Comments

**The science that informs the important threshold question in the review - what is the appropriate indicator or indicators? – needs to be fully vetted in the ISA. Thus, an explicit listing and discussion of potential indicators needs to be included in the ISA.**

While the choice of indicator is ultimately a policy decision, the ISA needs to adequately cover the science that would support that policy choice. Since a separate secondary standard with different indicator, form, averaging time, concentration, and/or allowed frequency of occurrence from the primary standard has not been in place for either SO<sub>x</sub> or NO<sub>x</sub>, the form and usefulness of alternative prospective indicators will be the important threshold question in the review. In fact, the December 2007 Review Plan indicates that the issue of appropriate indicators is central to the review of the standards.<sup>8</sup> The various options for indicator are not obvious from reading the draft ISA and so it is impossible to tell if it adequately covers the science behind each option. Therefore, an explicit listing and discussion of potential indicators needs to be included in the ISA. In CASAC comments on the draft review plan, members of the panel indicated that analysis of the many options for regulating deposition needs to be part of the ISA and requested that the technical foundations for use of potential indicators should be presented up front in the ISA.<sup>9</sup>

In addition to the potential indicators, there are also various policy options for how a secondary standard or standards utilizing the various indicators could be implemented under the Clean Air Act. Thus, the various technical options for protecting against acidification and eutrophication effects need to be stated up-front and then the data and tools needed to implement the various options need to be explicitly evaluated in the ISA. The state of knowledge (including uncertainties and limitations) for emission inventories

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<sup>7</sup> U. S. Environmental Protection Agency, Air Quality Criteria for Oxides of Nitrogen, EPA-600/8-91-048aF-cF, August 1993.

<sup>8</sup> U. S. Environmental Protection Agency, Integrated Review Plan for the Secondary National Ambient Air Quality standards for Nitrogen Dioxide and Sulfur Dioxide, December 2007.

<sup>9</sup> See CASAC letter to Administrator Johnson EPA-CASAC--08-003, November 29, 2007, at pages 31, 36, and 41.

(for both natural and man-made emissions of both reduced and oxidized forms of reactive nitrogen), atmospheric monitoring, atmospheric modeling, wet and dry deposition monitoring, and ecosystem monitoring and modeling need to be fully covered in the ISA.

**There are technical, legal, and policy considerations in choosing an appropriate indicator; the ISA needs to include a short discussion on the legal questions. For example, based on the Clean Air Act a strong case can be made that both primary and secondary national ambient air quality standards (NAAQS) must be nationally uniform.**

Section 109 of the Clean Air Act, which governs the establishment of NAAQS is entitled “National Primary and Secondary Ambient Air Quality Standards,” and Section 109 and its related provisions speak in terms of national standards, not regional or spatially different ones. Because the NAAQS provisions of the Clean Air Act have a national focus, its standard-setting process is not set up to allow EPA to distinguish between health or welfare effects in specific regions or geographic areas. In contrast, where Congress recognized a need for different levels of protection in different areas or regions, it specifically set up a regulatory framework to address the spatial differences, as in Title IV for acid deposition, in Section 112k for hazardous air pollutants in urban areas, and in Section 169a for visibility protection in federal class I areas (National Parks and Wilderness Areas). These specific provisions demonstrate that when Congress considered an issue with different spatial characteristics, it addressed the problem directly and expressly, not through the ambient air quality standard provisions of Section 109. Indeed, in over 30 years of applying the Act, EPA’s consistent practice has been to establish nationally uniform air quality standards.

The National Research Council study noted above concluded that concentration-based standards are inappropriate for protecting the resources discussed in the ISA. The study raised the option of adopting deposition-based secondary standards and possibly regionally distinct secondary standards. However, the study cautioned that a move to regional standards may require amendment of the Clean Air Act.<sup>10</sup>

**The ISA should evaluate both reduced and oxidized forms of reactive nitrogen, giving equal consideration to each. The discussion needs to start with a description and understanding of the natural N cycle.**

Since nitrogen is usually the limiting nutrient in terrestrial ecosystems, both the beneficial (through passive fertilization) and potentially adverse effects of nitrogen deposition need to be summarized and evaluated in the ISA. Since the deposition of reduced nitrogen can result in nitrate formation in soils via nitrification (the microbial-mediated reduction of the ammonium ion to the nitrate ion), both reduced and oxidized forms of reactive nitrogen need to be considered in acidification and eutrophication.

The ISA contains considerable discussion of what is referred to as the nitrogen cascade, in which man-made reactive nitrogen has increased globally due to (1) fossil fuel

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<sup>10</sup> NRC 2004 at page 312.

combustion, (2) synthetic fertilizer production, and (3) widespread production of crops (legumes, rice, etc.) that promote biological nitrogen fixation. The increased N cycled through the biosphere causes instances of nitrogen saturation that causes a variety of problems. While an increase in reactive nitrogen is not debated, the extent of the increase and the relative importance of reduced versus oxidized N forms should be discussed in the ISA. Such a discussion must start with the natural N cycle and what is known about the strength of various process and fluxes.

A key issue for interpreting the N and S deposition levels discussed in Chapter 4 of the ISA is the extent to which man's activities have perturbed the natural N and S cycles. In this regard, the recent paper by Luo et al.<sup>11</sup> which used a three-dimensional chemical transport model to evaluate the role of ammonia chemistry and natural aerosols on the global distribution of aerosols is informative. Among the findings Luo et al. report is that current levels of nitrate and ammonium deposition are twice pre-industrial levels while present sulfate deposition is almost five times pre-industrial levels. This finding should be referenced in the ISA.

**The beneficial effects of anthropogenic nitrogen deposition as it relates to fertilization/carbon sequestration/climate change needs to be evaluated in the ISA so it can be weighed in the policy considerations**

While acknowledging that increased reactive nitrogen causes both beneficial and adverse effects, the ISA focuses almost entirely on the adverse effects. However, one of the services that ecosystems provide is carbon sequestration.<sup>12</sup> For example, Gruber and Galloway,<sup>13</sup> in a recent publication, discuss the acceleration of the nitrogen cycle as a result of the production and use of nitrogen fertilizers. They note that a central question is how the availability of nitrogen will affect the capacity of the Earth's biosphere to continue absorbing carbon from the atmosphere and hence continue to help in mitigating climate change. Several CASAC panelists asked that climate change issues be included in the ISA and one provided references to papers that discuss the question of whether extra nitrogen input from man-made sources results in more carbon from the atmosphere being sequestered in forests.<sup>14</sup> While there are differing opinions in the literature on the magnitude of the effect,<sup>15</sup> there clearly is an effect and the extent of sequestration, as noted by Nadelhoffer et al., is determined by the portion of nitrogen that is present in woody and other tissues compared to the portion that is stored in soils, exported to aquatic systems, or released as nitrogen gases to the atmosphere.

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<sup>11</sup> C. Luo, C. Zender, H. Bian, and S. Metzger, "Role of ammonia chemistry and coarse mode aerosols in global climatological inorganic aerosol distributions," *Atmos. Environ.*, **41**, 2510-2543 (2007).

<sup>12</sup> ISA at page 4-2.

<sup>13</sup> N. Gruber and J. Galloway, "An Earth-system perspective of the global nitrogen cycle," *Nature*, **451**, 293-296 (2008).

<sup>14</sup> CASAC 08-003 at page 28.

<sup>15</sup> See P. Hogberg, "Nitrogen impacts on forest carbon," *Nature*, **447**, 781-782 (2007); F. Magnani, et al, "The human footprint in the carbon cycle of temperate and boreal forests," *Nature*, **447**, 848-450 (2007); K. Nadelhoffer, et al., "Nitrogen deposition makes a minor contribution to carbon sequestration in temperate forests," *Nature*, **398**, 145-148 (1999).

There is also a legal requirement that EPA consider the beneficial effects of an air pollutant as well as its adverse effects in both establishing criteria and setting NAAQS.<sup>16</sup> EPA acknowledges that it must assess the net impact on public health and/or welfare of a pollutant.<sup>17</sup> The draft ISA and any subsequent risk assessments, therefore, must provide a framework within which the net impacts of N deposition can be evaluated. The current draft does not adequately discuss either the beneficial effects of N or the possible tools that can be used to compare the benefits against the adverse effects.

**The issue of excess nitrogen or nitrogen saturation needs to be evaluated in relation to the natural N cycle**

Gruber and Galloway 2008 do this on the global scale. As noted above, the ISA should include a short description of the natural N cycle so that the more detailed description of man's perturbation of the natural cycle can be understood in perspective. The extent to which the fluxes from various processes and pathways (nitrification, denitrification, etc.) are known at the global and regional scales should be discussed. This information is relevant to the discussion in the ISA regarding the timeframe for nitrogen saturation, which is not known well and may be centuries rather than decades.<sup>18</sup>

**The level of detail in the ISA should be similar for both reduced and oxidized forms of reactive nitrogen**

For example, the kind of sources, spatial distribution of sources, trends in emissions, and potential effects of deposition are different between reduced and oxidized forms. The current draft focuses primarily on the oxidized forms. However, NO<sub>x</sub> emissions from fossil fuel combustion have been decreasing for several decades and will continue to decrease for the next several decades due to controls that are already "on the books." In contrast, the sources of reduced nitrogen (such as fertilizer use) are still increasing as noted below in the specific comments on ammonia emissions inventories. Therefore, additional detail on the sources and trends in reduced N emissions should be added and indicators that include reduced nitrogen should be explicitly evaluated. Several members of the CASAC panel, in comments on the draft review plan, agreed that reduced nitrogen needs to be fully included in the ISA.<sup>19</sup>

**Similarities and differences between sulfate and nitrate behavior and effects need to be clearly delineated.**

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<sup>16</sup> See March 6, 2008 Office of Management and Budget memo from Susan Dudley to Administrator Johnson at footnote 1, quoting the Court's decision in *American Trucking Association v. EPA* that legally EPA must consider positive identifiable effects of a pollutants presence in ambient air in formulating air quality criteria under section 108 and NAAQS under section 109 of the Clean Air Act.

<sup>17</sup> See March 7, 2008 U. S EPA memo from Marcus Peacock to Susan Dudley of OMB at page 2, indicating that EPA agrees that it must consider the beneficial effects of an air pollutant as well as its adverse effects, and that it must assess the net impact of a pollutant.

<sup>18</sup> ISA at page 4-43.

<sup>19</sup> See CASAC -08-003 at pages 4, 7, 17, 23, 36, and 41.

For example, sulfate is the dominant anion in surface water acidification over most of the U. S. and as S emissions have been reduced in recent decades, reductions in sulfate in surface water have been observed. In contrast, the contribution of nitrate to acidification varies substantially from region to region and the ISA indicates that there is little or no apparent relation between recent trends in N deposition and recent trends in nitrate in surface waters in the eastern U. S. In addition, the processes controlling and time frames for S and N saturation and recovery are different and need to be clearly delineated. The differences will become important as various potential indicators are evaluated in subsequent portions of the review.

**Basic information on land use patterns and changes in land use that may effect relevant ecosystems needs to be included in the ISA**

This is important because of the heterogeneity of ecosystems and heterogeneity of positive and negative effects that a given level of deposition may cause. For example, the same level of N deposition may be beneficial on agricultural lands, but potentially adverse on a nearby forest. The same level of deposition may be beneficial on another nearby forest that is being managed for lumber production. Thus, information on the extent and location of managed as well as unmanaged ecosystems needs to be included in the ISA.

Information on changes in land use are also important to put the effects studies into perspective. The ecosystem changes which occur with changes in land use that are routinely accepted in our society are often massive compared to the first effects of deposition on plant communities, lichen communities, fish stocks, etc. In addition, since ecosystems are constantly changing in response to natural variation in environmental stresses, competitive dynamics, and other natural processes, the definition of when a change becomes adverse is not straightforward. Sufficient background on these questions is needed in the ISA to inform the policy decisions which will be made later in the review.

**For all the major effects of concern, the ISA documents that the sensitivity of various ecosystems varies widely, making a national standard inappropriate.**

For acidification issues, the sensitivity of various ecosystems varies widely, based on bedrock geology, topography, soil chemistry, land use, and hydrologic flowpath. In coastal waters and estuaries, nitrogen inputs from atmospheric deposition, agricultural runoff, livestock waste, urban runoff, and wastewater discharge have led to eutrophication. The portion of reactive nitrogen from direct atmospheric deposition to the waterway and from deposition to the watershed varies substantially from place to place. In addition, the sensitivity of waterways depends on local conditions of morphology and water flushing dynamics.

## **Specific Comments**

### **Chapter 1 – Introduction**

Both the Review Plan (at page 3-2) and the ISA (at page 1-2) include lists of framing questions that the ISA and, ultimately, the current review must answer. The ISA, however, does not address these questions explicitly. Questions such as “Does the available information provide support for considering joint standards or are separate standards appropriate?” and “What forms of alternative standards are supported by the information and what are the uncertainties and limitations in that information?” are some of the questions that need to be explicitly addressed in the ISA. In section 1.1.2, the scientific basis for the administrator’s decision in the previous review should be summarized. As noted above, although there was substantial evidence of NO<sub>x</sub>-related acidification of freshwater streams and lakes and eutrophication of estuaries and coastal waters, the Administrator concluded that adoption of a nationally-uniform secondary standard would not be an effective way of approaching these problems.<sup>20</sup> The Administrator indicated that the decision would be re-visited in the next review and that, meanwhile, the Title IV program that addresses acid deposition and other national and regional programs will be reducing NO<sub>x</sub> emissions.

### **Chapter 2 – The Atmospheric Chemistry and Physics of Nitrogen and Sulfur Oxides**

#### **Clarity in the nomenclature is imperative to avoid confusion**

The introduction to Chapter 2 indicates that the ISA will use the term NO<sub>x</sub> to refer to all forms of oxidized nitrogen compounds in contrast to the nomenclature typically used in the scientific community wherein NO<sub>x</sub> refers to the total of just two of the oxides of nitrogen, NO and NO<sub>2</sub>. However, at numerous places in the ISA there is confusion since NO<sub>x</sub> is used as it used in the general literature. For example, the description of nitrogen chemistry at pages 2-3 to 2-7 uses the nomenclature where NO<sub>x</sub> + NO<sub>z</sub> = NO<sub>y</sub>, where NO<sub>y</sub> is the total of all oxidized nitrogen. To avoid such confusion, it is important to be very specific throughout the document to make sure that the chemical forms of nitrogen included in a given term or measured quantity are clear.

#### **The level of detail for reduced N should be the same as for oxidized N**

Chapter 2 notes that the ISA properly includes discussion of ammonia as a precursor of NO<sub>x</sub> in gas phase reactions as a source of ammonium that neutralizes acids as well as a separate actor in nitrification of terrestrial and aquatic ecosystems. Because of the importance of emissions, transport, transformations, and deposition of reduced forms of nitrogen, the ISA should include a review and discussion of reduced N throughout at the same level of detail as oxidized nitrogen.

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<sup>20</sup> 61 Federal Register 52852, October 8, 1996

### **Differences in the N deposition footprint between ground-level and elevated sources need to be discussed**

While the draft chapter includes several pages of description of prominent gaseous and heterogeneous reactions of NO<sub>x</sub> species, the reader does not get a good feel for the relevant time and spatial scales involved between emission and deposition. Since a knowledge of the spatial scale of influence (or footprint, as it might be called) of relevant emissions will be key input into the consideration of potential secondary standards, it is important to include additional discussion of the spatial scale. There are differences between the N deposition footprint due to NO<sub>x</sub> emissions emitted at ground level in urban areas generally in combination with VOC, and NO<sub>x</sub> emitted from tall stacks in combination with SO<sub>2</sub>, and reduced nitrogen emitted at ground level in rural areas.<sup>21</sup> For example, a recent study by Elliot, et al. 2007<sup>22</sup> of nitrogen isotopes in wet nitrate deposition reported that wet nitrate at 33 sites in the Midwestern and Northeastern U. S. is strongly associated with NO<sub>x</sub> emissions from stationary sources.

### **The description of atmospheric chemistry should focus on sources of non-linearity**

As indicated in Figures 2.2-1 and 2.3-1 and in text of Sections 2.2 and 2.3, the atmospheric chemistry of SO<sub>x</sub> and NO<sub>x</sub> is quite complex. For the purposes of the ISA, the draft should list and discuss the major sources of non-linearity in these coupled atmospheric oxidation processes. While NO<sub>x</sub> and SO<sub>x</sub> emissions that are emitted to the atmosphere will eventually be removed from the atmosphere, factors which will influence these processes in a non-linear way will shift where and when the materials are deposited, whether they are removed from the system or are re-cycled, and whether they are partitioned into different forms that may be more or less harmful or beneficial. Such differences are important and will influence the extent that current control programs reduce harmful deposition in sensitive areas and thereby affect potential adverse effects.

Chapter 2 includes discussion of heterogeneous chemistry involving halogen-radical reactions that may influence NO<sub>x</sub> and SO<sub>x</sub> oxidation especially in marine boundary layers. In the SO<sub>x</sub> section, it is acknowledged that the absolute magnitudes and relative importance of these processes are poorly understood. If this is true for the similar NO<sub>x</sub> reactions, it should be so stated. The extent to which the heterogeneous chemistry discussed in Chapter 2 is included in the atmospheric models discussed in Chapter 3 should be explicitly discussed.

### **The discussion of atmospheric measurements should acknowledge the limitations and drawbacks of current methods**

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<sup>21</sup> See also comment by CASAC panelist at page 44 of CASAC-08-003.

<sup>22</sup> E. Elliot, C. Kendall, S. Wankel, D. Burns, E. Boyer, K. Harlin, D. Bain, and T. Butler, "Nitrogen isotopes as indicators of NO<sub>x</sub> source contributions to atmospheric nitrate deposition across the Midwestern and Northeastern United States," *Environ. Sci. Technol.*, **41**, 7661-7667 (2007).

Chapter 2 also covers the atmospheric measurements of NO<sub>x</sub> and SO<sub>x</sub> species. The ISA acknowledges that routine NO<sub>2</sub> measurements in the monitoring network are subject to unknown and varying interferents from other nitrogen oxide species, so they are not a measure of either NO<sub>2</sub> or total oxidized nitrogen. Because of these concerns, there is a variable degree of positive bias in the data. The Health Criteria NO<sub>x</sub> ISA notes that this bias may be roughly 20 to 25 %.<sup>23</sup> Such a large positive bias is not acceptable. First, the specific techniques used to convert NO<sub>2</sub> to NO currently in use throughout the country should be documented by EPA and the States and the biases and interferences in those techniques should be carefully evaluated. Second, it is incumbent on the Agency to develop more specific NO<sub>2</sub> measurement techniques. The ISA also discusses research-type instruments or instruments under development that can measure NO<sub>y</sub> or individual NO<sub>x</sub> species noting issues and problems suggesting they are not ready for routine monitoring. Yet the ISA concludes that:

“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.”

This statement is too general and is not supported by the material in the ISA. In addition, a decision regarding adequacy is a policy decision; the ISA should stick to the facts concerning the monitoring methods. If concentration-based indicators are considered for secondary standards, there needs to be an explicit discussion of the techniques available for those indicators be they specific gaseous or particulate species or combinations and the possible utility of those indicators. For example, the ISA discusses measurement techniques for NO<sub>y</sub>, total oxidized nitrogen. However, the atmospheric concentration of total oxidized nitrogen would not be meaningful for evaluating ecological risk, since it a measure of a mixture of compounds which each have their own inherent deposition characteristics and possibly different effects in ecosystems. In addition, the atmospheric concentration of any species or combination of species would be an inappropriate indicator because it does not include wet deposition

### **The quality and completeness of deposition measurements should be discussed**

If deposition-based secondary standards are to be considered, a discussion of the quality and completeness of deposition measurements should be included in Chapter 2. Currently, there is no such discussion in the Chapter. Current measurements of dry deposition do not include gaseous ammonia and current measurements of wet and dry deposition do not include organic nitrogen compounds. In addition, dry deposition is not measured directly but calculated from atmospheric measurements of gaseous and particulate species. There are limitations and uncertainties in the current CASTNET procedures that are discussed in CASTNET reports.<sup>24</sup> These need to be discussed in the ISA. CASTNET is working to improve the set of measurement so there is also an issue

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<sup>23</sup> U. S. Environmental Protection Agency, First Draft Integrated Science Assessment (ISA) for Oxides of Nitrogen - Health Criteria, EPA/600/R-07/093, August 2007, at page 5-2.

<sup>24</sup> U. S. Environmental Protection Agency, Clean Air Status and Trends Network (CASTNET) 2006 Annual Report, November 2007.

of comparability as new measurements or procedures are instituted. This also needs to be acknowledged and discussed in the ISA.

### **Chapter 3 – Ecological Exposures to Oxides of Nitrogen and Sulfur, and to Ammonia and Ammonium**

**The coverage and discussion of oxidized nitrogen, reduced nitrogen and oxidized sulfur should be comparable. In particular, the discussion of nitrogen should be expanded while the discussion of sulfur should not be reduced.**

For example, the chapter discusses both trends in emissions and ambient measurements for SO<sub>x</sub> in Section 3.2 but not for NO<sub>x</sub> in Section 3.1. The reader should not have to go to a table in the annexes to find the breakdown of NO<sub>x</sub> or ammonia emission sources. The trends in reduced and oxidized N are also important to include in the body of the ISA.

To enable direct comparisons of various emission sources and fluxes in the N cycle, the quantities should be presented and compared on a consistent basis. Because of the potential use of N deposition as an indicator metric, the emission data should be reported on a mass of nitrogen basis. For example, NO<sub>x</sub> (NO + NO<sub>2</sub>) emissions are typically reported as mass of NO<sub>2</sub>. This results in a very misleading comparison between the mass of NO<sub>x</sub> and NH<sub>3</sub> emissions in Table AX2-1 when not presented on a mass of N basis.

In Section 3.3 the text indicates that the anthropogenic emissions of ammonia show a strikingly different pattern from that of NO<sub>x</sub> or SO<sub>x</sub>. However, that pattern is not shown. It should be. If emission trend data for reduced nitrogen is not available, trends in fertilizer use in the U. S. should be presented and discussed as a reasonable surrogate. For example, historic trends in fertilizer consumption in North America<sup>25</sup> and projections of nitrogen fertilizer demand in North America until 2011/2012<sup>26</sup> are available. In contrast to the continuing reductions in NO<sub>x</sub> emissions, N fertilizer demand in North America is forecast to increase at 0.3 % per year through 2012.

The text, in discussing NH<sub>x</sub> emission inventories, notes the Holland et al. 2005 study which compared NH<sub>x</sub> inventories to estimated deposition of reduced N species and found that ammonia emissions in the national emission inventory was underestimated by a factor of 2 to 3. This is an important finding that needs to be included in the discussion of uncertainty in inventories as well as in the overall integration of findings in subsequent chapters.

For both SO<sub>x</sub> and NO<sub>x</sub>, regulatory programs already in place are projected to reduce emissions well below current levels. The ISA should document these reductions since they will occur with or without separate secondary standards.

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<sup>25</sup>C. Gellings and K. Parmenter, Energy Efficiency in Fertilizer Production and Use, in Encyclopedia of Life Support Systems, developed by UNESCO, EOLSS publishers, Oxford, UK, 2004.

<sup>26</sup> Food and Agriculture Organization of the United Nations, Current world fertilizer trends and outlook to 2011/2012, Rome 2008.

**The discussion of NO<sub>x</sub> emissions from soil and lightning is missing important papers demonstrating that these sources result in significantly higher emissions than noted in the ISA**

There are NO<sub>x</sub> emissions from soils due to microbial processes, and fertilized agriculture produces the highest soil NO<sub>x</sub> emissions. The (Health) ISA references the Jaegle et al. 2005<sup>27</sup> study that used satellite observations of NO<sub>2</sub> columns to derive estimates of NO<sub>x</sub> emissions from fuel combustion, biomass burning, and soils. Jaegle et al. conclude that there is a significant role for soil NO<sub>x</sub> emissions at northern mid-latitudes during the summer, where they account for nearly half that of the fuel combustion source. The soil NO<sub>x</sub> estimated by Jaegle et al. was twice that in the inventory used in the GEOS-CHEM model. The Jaegle et al. estimate should be included in the ISA.

Another paper referenced in the ISA, Singh et al. 2007, found that lightning was a much larger contributor to NO<sub>x</sub> species in the upper troposphere over North America than previously thought. Singh et al. also refer to a new paper<sup>28</sup> in which the GEOS-CHEM model improved its prediction of the observational data in the upper troposphere by increasing the lightning source (NO<sub>x</sub> yield per flash) in the model by a factor of four. The Hudman et al. 2007 paper also provided boundary layer NO<sub>x</sub> measurements verifying a 50 % reduction in power plant and other NO<sub>x</sub> emissions over the eastern U. S. from 1999 to 2004. All these papers should be discussed in the ISA.

**The discussion of direct deposition to vegetation should include the fact that current levels of deposition directly to vegetation are not likely to cause risk or harm**

Section 3.5 discusses the uptake of N species to vegetation as an ecological sink. This section should include a reference to the fact that such interactions are not phytotoxic as noted in the conclusions on page 5-35. The complexities of these interactions is demonstrated by the fact that at low NO<sub>2</sub> concentrations (below 1 ppb) plants release NO<sub>2</sub> as documented in the material in Section 3.9.

**The material in Sections 3.6, 3.7, and 3.8 concerning monitoring needs to be expanded to provide more relevant information regarding monitoring of specific N species**

Section 3.6 on monitoring networks contains a significant amount of extraneous material covering monitoring networks for species not under consideration in the ISA. For the purposes of the ISA, the focus should be on networks such as IMPROVE, CASTNET, and NADP, due to their relevant measurement and locations. As noted above, Chapter 2 should include a detailed discussion of the measurement techniques for the dry and wet deposition measurements that are used in the ISA. The discussion of these networks is

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<sup>27</sup> L. Jaegle, L. Steinberger, R. Martin, and K. Chance, "Global partitioning of NO<sub>x</sub> sources using satellite observations: Relative roles of fossil fuel combustion, biomass burning, and soil emissions," *Faraday Discussions*, **130**, 407-423 (2005).

<sup>28</sup> R. Hudman et al., "Surface and lightning sources of nitrogen oxides over the United States: magnitudes, chemical evolution, and outflow," *J. Geophys. Res.*, **112**, D12S05 (2007).

sparse in Chapter 3; for example, the NADP network is noted as measuring “most” of the major ions key to aquatic chemistry addressing acidification and eutrophication effects.<sup>29</sup> To properly interpret the material in Chapter 4, the reader needs to know what is measured and what is not measured in each case, especially for measurements reported as “total” nitrogen. For example, the CASTNET maps in Section 3.8 are referred to in the text as total N but noted in the caption as total inorganic nitrogen. However, the data are actually neither since it does not include dry deposition from gaseous ammonia (as acknowledged in the section) or organic N which Section 3.11 notes can represent 30 % of rainwater N in deposition.<sup>30</sup>

Section 3.7 which discusses ambient concentrations should include the trend in ambient NO<sub>2</sub> as well as the summary of current levels that is included. For example, Figure 1 below shows the distribution of annual average concentrations at all U. S. monitoring locations from 1970 through 2007. While the number of monitoring sites differs somewhat from year to year, a downward trend that has reduced ambient NO<sub>2</sub> concentrations by a factor of about three over the past 30-some years is evident.

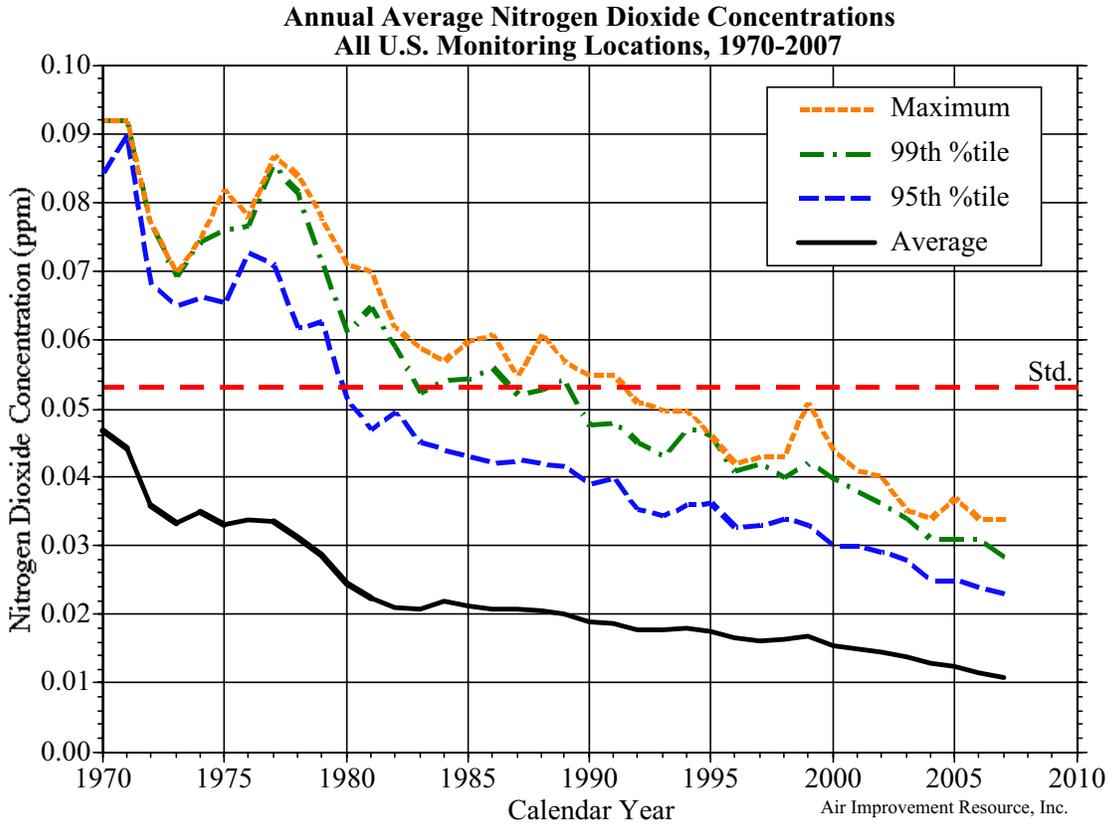
Since most of the NO<sub>2</sub> monitoring is conducted in urban or suburban locations, the ambient trend is primarily indicative of emission reductions in and around urban areas. Since highway vehicle emissions are a major source of NO<sub>x</sub> in urban areas, a comparison of the ambient trend with the trend in highway vehicle emissions is illustrative. Figure 2 shows the trend in highway vehicle emissions on a gram per mile basis from 1970 through 2020 calculated with the U. S. EPA’s MOBILE6 emission model, using the model’s default inputs. The trend in NO<sub>x</sub> emissions from highway vehicles from 1970 to 2007 is very similar to the trend in ambient NO<sub>2</sub>. Moreover, the downward trend in NO<sub>x</sub> emissions will continue well into the 2020s.

The data in Figures 1 and 2 provide strong evidence that (1) N deposition in and around urban areas has been reduced substantially since 1970, and (2) that it will continue to fall for the next several decades as the current fleet of highway cars and trucks is replaced with new, low-emitting vehicles under the current Federal motor vehicle control program.

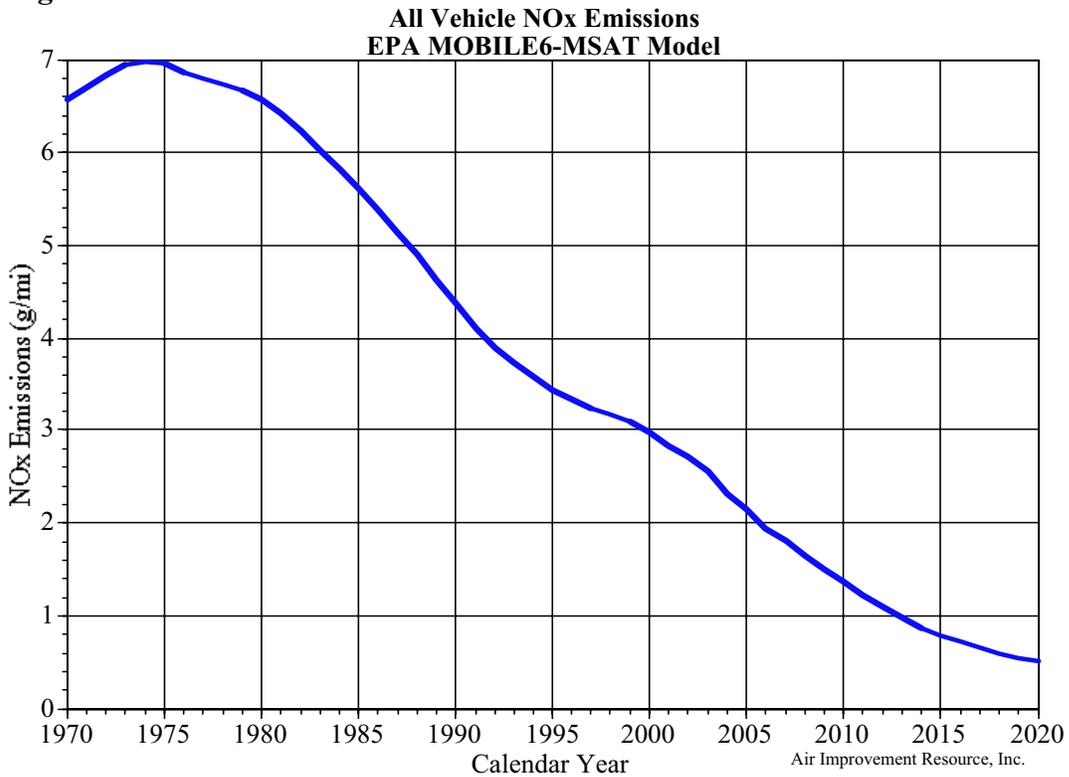
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<sup>29</sup> ISA at page 3-20.

<sup>30</sup> ISA at page 3-50.



**Figure 1**



**Figure 2**

The other large source of NO<sub>x</sub> emissions is power plants. These sources have also been subject to substantial control and are scheduled for additional control under the Clean Air Interstate Rule. The ISA should document the reductions that have taken place to date and the reductions that are anticipated with current regulations for both motor vehicle and power plant emissions in the ISA to provide context for the interpretation of historic environmental data

Regarding deposition, the maps in Section 3.8 from CASTNET are useful, but for the purposes of the ISA, additional detail should be provided. There should be separate displays of the spatial distribution and trends in oxidized N and reduced N deposition. This would provide the reader perspective on the overall footprint from reduced N and oxidized N emissions and enable a comparison with their respective emission trends. Because of the importance of base cations to the acidification issue, there should also be a discussion of the available information on levels and trends in deposition of base cations.

**It is not clear what Section 3.9 adds to the discussion**

Section 3.9 is a detailed discussion of the fluxes of NO<sub>x</sub> species in Harvard Forest. It is not clear why this much detail is needed in the body of the ISA. The bottom line from this work is not clearly stated in the section. It does demonstrate the complexities of N interactions in the forest canopy and ecosystem. It also demonstrates that vegetation can be both a source of NO<sub>2</sub> at low ambient NO<sub>2</sub> concentrations and a sink for N species. The text indicates that direct NO<sub>2</sub> by plants is important to the plant but not a significant part of the overall N deposition to rural sites

These complex interactions do, however, raise other questions which are not directly addressed in the ISA. There is the general question of the relative importance of direct deposition to vegetation versus deposition to soils or other aquatic resources in various ecosystems. There is also the question of the relative portion of and importance of deposition to managed ecosystems versus deposition to unmanaged ecosystems versus deposition to the built environment. There is also a question of the representative-ness of the CASTNET measurements which are generally made in open locations to the situation in a forest where the materials interact with the vegetation (and possibly the biogenic emissions from the vegetation) substantially before depositing on the forest floor. Section 3.9 should be re-cast to address the questions noted above and to summarize the complex interactions with vegetation that are presented in more detail in the Annex.

**The discussion of atmospheric modeling in Section 3.10 and 3.11 is too general. The focus should be on the performance of the CMAQ model (or other models) that will be used in the risk assessment**

Since the text indicates that most modeling efforts within EPA use CMAQ, the ISA should include an evaluation of that model's performance for applications relevant to the current secondary standard review. For example, one of the CASAC panelists points out that the heavy reliance on CMAQ necessitates the validation and improvement of that

model.<sup>31</sup> He further cautions that the marginal performance for non-sulfur species suggests considerable room for improvement. There are a number of recent papers that document the performance, including Eder and Yu.<sup>32</sup> Eder and Yu report that while the performance for sulfate is good, the performance for nitrate is relatively poor. In addition, the overall nitrate performance masks systematic over- and under-prediction problems that demonstrate substantial and compensating temporal and spatial differences in performance. In addition, Davis and Swall<sup>33</sup> document issues with the CMAQ predictions of the wet deposition of ammonium. Currently, the ISA references but does not discuss some CMAQ performance evaluations. Instead, it should carefully document and discuss all relevant CMAQ papers so that the technical limitations of CMAQ or any other model that will be used in the risk assessment are included.

The major comparison in the section is from the Detener et al. 2006b analysis of 23 global models. It is not clear what a comparison of mean model results with observations tell us and it is not clear how to interpret the conclusion that 60 to 70 % of the participating models capture the measurements to within 50 % in regions with quality controlled observations. Since global models will likely not be used in the risk assessment, the focus should be on regional and local atmospheric models and their performance on relevant metrics.

The text refers to Table 3.11-2 which is just a list of sources in various categories to support the conclusion that NO<sub>x</sub> chiefly from fossil fuel combustion often dominates N pollution in the United States and comprises about 50 to 75 % of total N atmospheric deposition. The current draft ISA does not contain enough of the supporting data to determine whether this conclusion is warranted or not, given the contributions from fertilizer use and natural sources. The basis for the conclusion should be explicitly provided in the ISA. With the inclusion of the data suggested in these comments, the reader can make his or her own evaluation of the contribution of oxidized versus reduced nitrogen and the contribution of fossil fuel combustion to overall oxidized nitrogen deposition.

**Although the policy relevant background concentration of NO<sub>2</sub> is far below current urban levels, an accurate definition and analysis of policy relevant background is necessary in the ISA.**

The ISA defines a policy relevant background (PRB)<sup>34</sup> in Section 3.12 as those concentrations that would result in the United States in the absence of anthropogenic emissions in North America; i.e. leaving no room for NO<sub>2</sub> caused from human activities. This definition of PRB is flawed for several reasons. It omits the contribution from biogenic emissions due to agricultural activities and it omits consideration of the contribution from anthropogenic emissions in Mexico and Canada. While there can be

<sup>31</sup> CASAC 08-003 at page 27.

<sup>32</sup> B. Eder and S. Yu, "A performance evaluation of the 2004 release of Models-3 CMAQ," *Atmos. Environ.*, **40**, 4811-4824 (2006).

<sup>33</sup> J. Davis and J. Swall, "An examination of the CMAQ simulations of the wet deposition of ammonium from a Bayesian perspective, *Atmos. Environ.*, **40**, 4562-4573 (2006).

<sup>34</sup> ISA at page 3-51.

reductions in biogenic emissions from agricultural activities, as with other human activities, it would be impossible to drive them to zero. No-one contemplates the cessation of human activities including the practice of growing crops and raising animals in the U.S. Omitting the impact of Canadian and Mexican emissions is also problematic. The U.S. may have treaties with these countries, but achieving zero anthropogenic emissions by ceasing human activities is both infeasible and impossible to be controlled by U.S. regulations.

The ISA also errs by relying on one model rather than considering a range of results from both models and measurements. Although background is not as large an issue in the NO<sub>x</sub> secondary standard review as it is in the ozone review, studies included in the ISA show that EPA systematically underestimated natural NO<sub>x</sub> emissions and, hence, both background NO<sub>2</sub> in this review and background ozone in the ozone review. For example, the ISA indicates that the model used to estimate PRB for NO<sub>2</sub> (Mozart-2) has lower estimates (by about a factor of two) of soil NO<sub>x</sub> than the GEOS-CHEM model that was used to estimate PRB for ozone.<sup>35</sup> As noted above, the (Health) ISA references the Jaegle et al. 2005<sup>36</sup> study that used satellite observations of NO<sub>2</sub> columns to derive estimates of NO<sub>x</sub> emissions from fuel combustion, biomass burning, and soils. Jaegle et al. conclude that there is a significant role for soil NO<sub>x</sub> emissions at northern mid-latitudes during summer, with estimated soil NO<sub>x</sub> emissions twice that in the inventory used in the GEOS-CHEM model. Thus, the Mozart-2 model used in the ISA likely underestimates soil NO<sub>x</sub> at northern mid-latitudes by roughly a factor of four.

By turning off the biogenic agriculture source in the PRB analyses, the Agency systematically underestimated uncontrollable background in both the NO<sub>2</sub> and ozone reviews. As noted above, the Singh et al. 2007 and Hudman et al. 2007 papers found that lightning was a much larger contributor to NO<sub>x</sub> species in the upper troposphere over North America than previously thought, by about a factor of four. This also results in a systematic under-prediction of background concentrations and deposition.

### **The Summary of Chapter 3 requires revision**

The summary should document current levels and trends for the relevant species and metrics. As noted above, it does for some species and metrics but not others. It correctly notes that ambient annual NO<sub>2</sub> concentrations have decreased 35 % from 1990-2005 and ambient annual SO<sub>x</sub> concentrations have decreased 50 % from 1990 to 2005. Figures 1 and 2 document that these trends are part of a longer trend that will continue as highway vehicle and power plant emissions continue to decrease under current control programs. The summary discusses the importance of ammonia from agriculture noting increased emissions over recent decades but does not provide a quantitative estimate of the trend. It summarizes current deposition patterns but not trends.

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<sup>35</sup> ISA at page 3-54.

<sup>36</sup> L. Jaegle, L. Steinberger, R. Martin, and K. Chance, "Global partitioning of NO<sub>x</sub> sources using satellite observations: Relative roles of fossil fuel combustion, biomass burning, and soil emissions," *Faraday Discussions*, **130**, 407-423 (2005).

## **Chapter 4 – Effects of Acidification and Nitrogen Enrichment on Ecosystems and other Welfare Effects**

### **Section 4.1 – Introduction to Ecological Concepts**

The introduction to Chapter 4 notes that Section 4.1 frames several important concepts for the assessment including the challenges of defining adversity. In particular, Section 4.1.3 notes that quantifying adverse effects on ecosystems has been a challenge to the secondary NAAQS process from the onset and mentions two approaches. The Critical Loads approach is described in Section 4.1.3.1 and in Appendix 8, while the valuation approach is discussed in Appendix 10. Because of the central importance of how adversity is defined and evaluated in the review, these two concepts need to be fully discussed in the body of the ISA. In particular, the question of how either concept could be incorporated into and implemented in the NAAQS process should be addressed.

#### **Issues with the valuation approach need to be explicitly discussed in the body of the ISA**

The evaluation approach inherently involves a cost-benefit assessment. For example, a recent National Research Council report recommends that “policymakers should use economic evaluation as a means of evaluating the trade-offs involved in environmental policy choices; that is, an assessment of benefits and costs should be part of the information set available to policymakers in choosing among alternatives.”<sup>37</sup> For nitrogen deposition on ecosystems, the ISA indicates that there are both benefits and adverse effects. Thus, the balancing of effects should consider both positive and negative impacts of nitrogen deposition as well as the costs of control.

Whether data exists to implement a valuation approach is also an important issue. In the Regulatory Impact Analysis for the Clean Air Interstate Rule, EPA discussed the impact of the emissions reductions in the rule on ecosystems and the fact that there are both positive (passive fertilization) and negative (acidification and eutrophication) effects from nitrogen oxides emissions, but indicated that the deposition benefits from the rule could not be monetized<sup>38</sup> Chapter 10 of the Annex discusses the lack of data and detailed understanding of deposition impacts that limit the ability to monetize benefits.

#### **Issues with the critical loads approach need to be explicitly discussed in the body of the ISA**

The Critical Loads approach is another framework that can be used for defining adversity. However, as noted in the ISA, there is no single definitive critical load for a natural resource and, the inescapable heterogeneity of ecosystems, together with the multitude of policy choices involved in developing critical loads complicates the analysis.

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<sup>37</sup> National Research Council, *Valuing Ecosystem Services: Towards Better Environmental Decision-Making*, National Academies Press, Washington, DC, 2004, at page ES-6.

<sup>38</sup> U. S. Environmental Protection Agency, *Regulatory Impact Analysis for the Final Clean Air Interstate Rule*, EPA 452/R-05-002, March 2005

When first introduced as a concept, the critical load was viewed as a threshold below which effects do not occur and above which they do. However, as the concept has been developed and applied, the situation has become much more complex due to uncertainties throughout the chain of information needed to apply the concept. Therefore, the ISA did not define critical loads for the U. S.

However, the ISA does include considerable discussion of the critical loads approach in Chapter 8 of the Annex. That discussion is general in nature and appears to be taken from European documents since it uses words like “protocol” and “negotiation.” Chapter 8 does point out several places in the U. S. where critical loads are being evaluated cooperatively – among states and between the U. S. and Canada. It also notes that the approach has been used extensively in Europe for organizing information about effects and for specifying emission reductions that would be required to protect ecosystems.<sup>39</sup>

### **The European experience with critical loads is informative and should be discussed in the ISA**

The ISA should summarize the nature and status of the use of critical loads in Europe in some detail because that experience can inform the potential use of critical loads in the U. S. as it relates to secondary standards. The critical loads concept is being used as part of the multi-country negotiations under the United Nations Convention on Long-range Transboundary Air Pollution (LRTAP). Since the size of European nations is akin to the size of states in the U. S., an international mechanism was needed to address the problem of emissions that arise in some countries being transported to, deposited in, and having effects in downwind countries. The LRTAP Convention was put in place to provide an international forum for analyzing and solving such transboundary pollution issues. The U. S. and Canada are also parties to the Convention.

One of the Protocols under the Convention, is the 1999 Gothenburg Protocol to Abate Acidification, Eutrophication and Ground-level Ozone. The Protocol sets emission ceilings for 2010 for four pollutants, sulfur, NO<sub>x</sub>, VOCs, and ammonia. The ceilings, if fully implemented, would reduce Europe’s sulfur emissions by 63 % compared to 1990 levels. For NO<sub>x</sub>, the ceiling represents 41 % control, and for ammonia 17 % control from 1990 levels. These ceilings were negotiated on the basis of scientific assessments of pollution effects and abatement options. They do not represent the emission reductions required to attain critical loads (or critical levels of ozone) throughout the region, although they are expected to reduce the area of Europe with excessive levels of acidification and eutrophication substantially.

Although the Protocol was signed in 1999, it did not go into effect until 2005 when a sufficient number of countries had signed onto their obligations under the Protocol. Under the LRTAP Convention, the parties (countries) are in the process of evaluating further reductions beyond 2010 using a critical loads framework. A November 2007

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<sup>39</sup> ISA at page AX8-2.

progress report<sup>40</sup> indicates that there are many simplifications and data limitations in the effort to model N critical loads. In addition, the Europeans are evaluating emission reductions against two sets of critical loads, one based on modeling and a second they refer to as expert-based empirical critical loads. The empirical critical loads are provided in Table AX8.4-3. They are given as a range, because of variation in sensitivity within a given ecosystem. In addition, the Europeans have designated some of these critical loads as “reliable”, some as “quite reliable”, and some as based on “expert judgement,” when there is little or no data. The two sets of critical loads are substantially different from each other. This preliminary European effort is evaluating the impact in 2020 of a “maximum feasible control” scenario versus the current legislation baseline. Thus, the Europeans are using the critical loads concept together with atmospheric modeling to evaluate the cost and effectiveness of various control options. These analyses, when sufficiently refined, will then be input into a political process by which the countries negotiate the next round of control requirements.

As noted above, the critical load concept was originally viewed as a threshold below which effects do not occur and above which they do. However, as the concept has been developed and applied in Europe and elsewhere, the situation has become much more complex due to uncertainties throughout the chain of information needed to apply the concept. In the United Kingdom, uncertainties in critical loads have been extensively evaluated.<sup>41</sup> Because of the wide heterogeneity in ecosystems, much of the critical loads information is presented in maps to show local variations in sensitivity. However, the UK evaluation concluded that it is inadvisable to attempt to interpret national scale maps of critical loads at a local or site-specific scale. Since national scale in the UK is equivalent to state-scale in the U. S., this means that even state-scale information cannot be used to accurately identify critical loads at a specific site. Because of all the uncertainties involved, the Europeans present results in terms of probability distributions. The UK report concludes that at no site can we be 100 % confidence of either exceedance or non-exceedance of a critical load.

Although the Europeans have been developing critical loads as a tool for evaluating adversity and setting policy, they do it in a cost-benefit framework. This is not consistent with the U. S. NAAQS paradigm. In addition, the Europeans have not had any greater success in actually reducing emissions than has the U. S. under the mix of control approaches included in the U. S. Clean Air Act and individual state clean air authorities.

## **Section 4.2 - Ecological Effects of Acidification**

### **Similarities and differences between sulfate and nitrate need to be highlighted**

While the bulk of the material in Chapter 4 is a reasonable summary of the existing information, for the purposes of integrating the information and making the choices later

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<sup>40</sup> J.-P. Hattelingh et al., Status of European critical loads with emphasis on nitrogen, Chapter 1 in Coordination Centre for Effects (CCE) Progress Report 2007, Bilthoven.

<sup>41</sup> R. Skeffington et al., Uncertainty in critical loads assessment models, Science Report SC030172/SR, The Environment Agency, United Kingdom, March 2007.

in the review process, a number of the similarities and differences between the behavior of sulfate and nitrate need to be highlighted. While both sulfate and nitrate can acidify ecosystems, there are important differences. Sulfate in excess of biological demand has occurred for many decades in portions of the U. S. so sulfate leaching into surface water has been the primary cause of acidification effects.<sup>42</sup> Over recent years SO<sub>x</sub> emissions have been reduced dramatically and even further reductions will occur with regulatory programs that are already in place. As a result of the reductions to date in emissions, there have been documented reductions in sulfate deposition and variable amounts of recovery from the effects of acid deposition in various ecosystems around the country. The main issue for SO<sub>x</sub> in the review is the presence of accumulated sulfur stored in soils and the dynamics of storage and release and recovery as SO<sub>x</sub> emissions continue to decrease.

For nitrate, the situation is different. First, the ISA notes that in agricultural areas and especially in forested areas it is generally expected that most atmospherically deposited N is taken up by terrestrial vegetation.<sup>43</sup> This is an important distinction. Nitrogen is usually a limiting nutrient for terrestrial ecosystems.

Second, the cycling of N in ecosystems is much more complex than for S and is dominated by biological processes. The ISA notes that N deposition causes acidification by two mechanisms – (1) excess accumulation in soils followed by increased rates of nitrification by microbes and (2) change in base cation status of soils caused by nitrate leaching. Nitrification is the microbial-mediated reduction of the ammonium ion to the nitrate ion. Thus, the deposition of reduced nitrogen can result in nitrate formation in soils. There is also denitrification whereby bacteria reduce oxidized nitrogen to nitrogen gas and mineralization in which organic nitrogen is converted by microorganisms into plant-available inorganic forms. Thus, as acknowledged in the Annexes, most nitrate leaching is under biological control.<sup>44</sup> Thus, N dynamics is quite different from that of S.<sup>45</sup>

Third, surface water nitrate is generally much lower and more variable than surface water sulfate.<sup>46</sup> Nitrate leaching also tends to be more episodic, associated with snow melt and rainstorms. Even so, nitrate that is leached has been cycled by biota in soils or streams.<sup>47</sup>

Fourth, trends in N deposition and its potential effects are less clear and the timescale of N saturation may be longer than for S and longer than previously thought. During the 1990s, total N deposition was relatively constant, with NO<sub>x</sub> emissions decreasing and reduced N emissions increasing over the decade. The ISA notes that during the 1990s the relationship between N deposition and nitrate in surface water was complex with different trends in different areas.<sup>48</sup> Efforts to explain the inconsistent trends were

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<sup>42</sup> ISA at page 4-8.

<sup>43</sup> ISA at page 4-13.

<sup>44</sup> ISA at pages AX4-30 and AX5-11.

<sup>45</sup> ISA at page 4-82.

<sup>46</sup> ISA at page 4-40.

<sup>47</sup> ISA at page AX5-11.

<sup>48</sup> ISA at page 4-41.

focused on both terrestrial and aquatic N cycling, reflecting the complexities of N utilization within terrestrial and aquatic ecosystems<sup>49</sup> The ISA also notes that the time scale of N saturation may be longer than previously considered (centuries rather than decades).<sup>50</sup>

Fifth, the portion of forest and/or aquatic systems that are affected is relatively small and we do not have a good feel for how that portion is changing. There is widespread agreement as to the ecosystems that are most sensitive to acidification, lakes and streams at relatively high elevation in areas that have base-poor bedrock, high relief, and shallow soils. The principal factor governing sensitivity is geology, followed by topography, soil chemistry, land use, and hydrologic flow path. While there are a number of areas with detailed research evaluating N and S budgets and trends and a number of surveys conducted over the years to identify the portion of resources affected, the surveys are difficult to compare since they used different approaches and sampling strategies. Another limitation is that there have been no studies in the U. S. to determine if the severity or frequency of episodic acidification has changed in response to the reductions in acid deposition over the past 30 years.<sup>51</sup>

Sixth, man-made and natural disturbances affect N cycling and the response of forests to N deposition. The ISA acknowledges that timber harvesting road-building, agricultural development, fire, drought, and storm damage can all impact N utilization and nitrate leaching.<sup>52</sup>

Seventh, the mechanisms of retention and release are not fully understood for N making ecosystem modeling more difficult for N than for S. The ISA needs to explicitly discuss the differences in N and S modeling and the current state of science for this aspect of ecosystems.

Eighth, although there a large number of studies of the impact of N additions to forested and other ecosystems, with almost all showing fertilization and growth, the effects have been wide ranging as to when positive effects turn to negative effects.<sup>53</sup> For the ISA, the issue is not the effect of increasing N deposition but the effect of reducing it from current levels by various amounts.

### **Potential indicators and forms of potential standard(s) need to be explicitly identified and discussed**

As the ISA integrates the various findings and discusses potential indicators for a secondary standard or standards, the eight factors or differences noted above need to be considered.

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<sup>49</sup> ISA at page 4-43.

<sup>50</sup> Ibid.

<sup>51</sup> ISA at page 4-22.

<sup>52</sup> ISA at page 4-125.

<sup>53</sup> ISA at page 4-73.

A key question/issue is at what level of a potential indicator do effects appear and what the level of that indicator will be in the future as NO<sub>x</sub> and SO<sub>x</sub> reductions already “on the books” are implemented. The ISA should address the first part of this question and the risk assessment should address the second.

Another key issue is how to handle the vast difference in ecosystem sensitivity from place to place. The framework established for national air quality standards by Congress did not envision such regional or local differences. Although establishing secondary air quality standards is one potential option for dealing with acidification and eutrophication effects, it is not the only option. Indeed, there already are numerous examples of cooperative regional efforts to understand and deal with acidification and eutrophication.<sup>54</sup>

### **Section 4.3 – Nitrogen Nutrient Addition**

This section discusses nutrient nitrogen addition within the framework noted above of the nitrogen cascade, in which man-made reactive nitrogen has increased globally due to (1) fossil fuel combustion, (2) synthetic fertilizer production, and (3) widespread production of crops (legumes, rice, etc.) that promote biological nitrogen fixation. The increased N cycled through the biosphere causes instances of nitrogen saturation that causes a variety of problems. Since nitrogen is usually the limiting nutrient in terrestrial ecosystems, the increased cycling of N has both beneficial (through passive fertilization) and potentially adverse effects.

Section 4.3.2.1 introduces the description of nitrogen saturation first given by Aber et al. 1989 which describes the process of response to long-term chronic N additions in four stages. In Stage 0, N inputs are low and there are strong N limitations on growth. Stage 1 is characterized by high N retention and a fertilization effect of added N on tree growth. Stage 2 includes the induction of nitrification and some nitrate leaching although growth may still be high. In Stage 3 tree growth declines and nitrification and nitrate loss continue to increase.

The ISA defines nitrogen saturation as the condition whereby the inputs of N to the ecosystem exceed the requirements of terrestrial biota, and consequently an elevated fraction of incoming N leaches out of soils to surface waters. This definition is different from that first offered by Aber et al. 1989. Instead of exceeding the requirements of terrestrial biota, Aber et al. used the phrase “exceeds biotic uptake capacity.” This is an important difference. The thought that each ecosystem has some fixed requirement for N implies a steady state and that every change is somehow unnatural and adverse. In contrast, ecosystems are dynamic, competitive systems subject to many stresses including pollution. In managed ecosystems, we use fertilization and other techniques to increase

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<sup>54</sup> For example, Chapter 3 of the Great Waters Third Report to Congress summarizes numerous national control programs that are reducing NO<sub>x</sub> emissions and numerous state, local and regional programs that are addressing specific estuaries and their pollution problems around the country. In addition, the Final Report of the Multi-Agency Critical Loads Workshop held in May 2006 summarizes a number of programs underway to apply critical loads in specific regions and ecosystems in the U. S. as a tool to understand and manage sulfur and nitrogen deposition issues.

food production or provide for more useful or aesthetically pleasing environments. These are benefits from the increased N cycling. The main issue for society is how to manage the sources of reactive N to maximize the benefits while minimizing the adverse effects of nitrate leaching. The ISA, while focusing on the potentially adverse effects of excess N, should include an evaluation of the benefits of N and the need for a balanced solution.

Since there are emissions of both oxidized and reduced forms of N involved in N nutrient issues, the ISA should specifically address any data available to evaluate whether the chemical form of the N is important in specific ecosystems. For example, the form of added N is important for wetlands<sup>55</sup>

### **The potential role of secondary NAAQS standards versus other regulatory approaches needs to be discussed for the most sensitive ecosystems across the U. S.**

The discussion of most sensitive and most affected ecosystems in Section 4.3.4 indicates that in the western U. S. there is concern for alpine ecosystems in the Colorado Front Range, chaparral watersheds of the Sierra Nevada, and Southern California sage shrub communities. While there have been high NO<sub>x</sub> emissions and deposition historically in the Los Angeles Basin and San Joaquin Valley of California, there have been major reductions in NO<sub>x</sub> emissions over the past several decades and the State Implementation Plans for these areas to meet the current national ozone standard require drastic additional cuts in NO<sub>x</sub> emissions. Therefore, a national secondary standard is not needed to address NO<sub>x</sub> effects in these areas of California.

For the Colorado Front Range alpine ecosystems, the ISA indicates that two specific power plant's emissions are involved<sup>56</sup> If so, local regulations are a more appropriate regulatory tool than a separate national secondary standard.

In the eastern U. S., the ISA uses Stoddard's 1994 estimate of the distribution of watersheds in different stages of N saturation. Of 85 northeastern watershed, 40 % were in Stage 0, 52 % in Stage 1 and 8 % in Stage 2.<sup>57</sup> Those in Stage 1 or 2 were most prevalent in the Adirondack and Catskill Mountains. Driscoll et al. 2007 reports that many lakes in the Adirondacks had decreases in nitrate over the period 1992-2004 and that the pattern of decreasing nitrate runs counter to what would be expected if the Adirondacks were approaching a condition of N saturation.

The ISA also notes a series of papers reporting locations where lake or streamwater nitrate has been reported. However, it also notes that all the regions except Colorado experienced N deposition greater than 10 kg N ha<sup>-1</sup> yr<sup>-1</sup> through the 1980s and 1990s.<sup>58</sup> This is consistent with the results of Aber et al. 2003 which associated the onset of nitrate leaching in eastern forests with deposition levels of 10 kg N ha<sup>-1</sup> yr<sup>-1</sup> in a compilation of data from surveys of 354 sensitive lakes and streams in 1990s.

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<sup>55</sup> ISA at page 4-133.

<sup>56</sup> ISA at page 4-149.

<sup>57</sup> ISA at page 4-148.

<sup>58</sup> ISA at page 4-151.

Current deposition of inorganic N from the monitoring sites noted in Chapter 3 is the order of  $10 \text{ kg N ha}^{-1} \text{ yr}^{-1}$  or less as shown in Figure 3.8-3. There will be major reductions in NO<sub>x</sub> emissions from power plants in the eastern U. S. due to the CAIR rule (a 61 % reduction from 2003 levels by 2015 in the CAIR region) and nation-wide from the ongoing federal motor vehicle control program throughout the U. S. Therefore, the oxidized N contribution to deposition will decrease substantially in coming years due to controls already on the books.

### **The implications of the fact that there are many sources that contribute to eutrophication need to be discussed**

Eutrophication of estuaries and coastal waters is another major concern related to N nutrient addition. Eutrophication is a condition in which a body of water becomes over-rich in nutrients and results in nuisance or toxic algal blooms and reduced oxygen availability. In the U. S., as described in sections 4.2.3.3 and 4.3, this is a special concern in estuaries and marine ecosystems. Eutrophication is not primarily an air pollution-related problem. It is a matter of both water and air pollution, with N additions from both atmospheric deposition to watersheds, direct deposition to water surfaces, wastewater discharge, agricultural runoff and urban runoff. It therefore involves not only combustion sources of NO<sub>x</sub>, but also agricultural practices, fertilizer use, treatment of human waste, and treatment of animal waste. The ISA summarizes several studies of the national extent of eutrophication (Bricker et al. 2007) as well as studies of individual watersheds. There is a wide range of findings for the extent of eutrophication and the contribution of atmospheric deposition to specific water bodies. The ISA notes that the estimates of atmospheric contribution entail considerable uncertainty since they depend on many assumptions.<sup>59</sup> The wide range of atmospheric contributions, the wide range of sensitivities of different water resources, and the wide range of contributions from non-atmospheric sources makes using a national standard to address the situation problematic. The ISA should acknowledge that secondary air quality standards cannot, by themselves, solve the nation's eutrophication problems and that the atmospheric contribution of oxidized N to eutrophication is being substantially reduced under current control programs.

There are a wide range of existing regulatory frameworks that are already addressing the eutrophication of estuaries and other waters in the U. S. For example, the Chesapeake Bay Program initiated in 1983 by the EPA and states surrounding the Bay is discussed in the ISA.<sup>60</sup> The Great Waters Report to Congress<sup>61</sup> contains a long list of research and regulatory programs that affect N and other water pollution issues in the country. All these programs at the federal, state, and local levels have been focused on reducing pollutant loads without the existence of separate national secondary standards for SO<sub>x</sub> and NO<sub>x</sub>. Thus, a great deal of regulatory tools and flexibility already exist to allow

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<sup>59</sup> ISA at page 4-121.

<sup>60</sup> ISA at page 4-161.

<sup>61</sup> U. S. Environmental Protection Agency, Deposition of Air Pollution to the Great Waters: Third Report to Congress, EPA453/R-05-005, June 2000.

cooperative efforts to study and solve the nation's eutrophication problems. The ISA should acknowledge and enumerate all the different existing regulatory authorities and programs that affect eutrophication.

## **Chapter 5 – Findings and Conclusions**

The chapter is more of a list of findings from the literature than an integrated synthesis of the information that would be the basis for consideration of separate secondary standards. There is no specific discussion of potential indicators although the conclusions section, 5.7, does use the metrics of  $\text{kg N ha}^{-1} \text{ yr}^{-1}$  and  $\text{kg S ha}^{-1} \text{ yr}^{-1}$ . For example, reference is made to modeling studies of Shenandoah National Park indicating that re-acidification may be prevented if deposition is kept between  $9\text{-}15 \text{ kg N ha}^{-1} \text{ yr}^{-1}$  and  $0\text{-}6 \text{ kg S ha}^{-1} \text{ yr}^{-1}$ . In addition, reference is made to several different levels of N deposition that have been associated with effects in different ecosystems and parts of the country.

There is a general statement that “Overall, we conclude that there is a causal relationship between current levels of S and N deposition and numerous biologically adverse effects on ecosystems in the United States.” Since the strongest evidence of ecosystem effects comes from locations that have had high historic deposition levels and since deposition levels of S and oxidized N have been decreasing, it is not so clear that current levels of deposition are causing all the adverse effects noted in the ISA. More importantly, since levels of S and  $\text{NO}_x$  deposition will be decreasing substantially over the next several decades under current control programs, it is not clear that additional control of combustion  $\text{NO}_x$  and  $\text{SO}_x$  beyond that which will occur under existing regulatory programs is needed.

There is also a paragraph that indicates that the current regulatory networks for S and N deposition are inadequate to characterize heterogeneity or hotspots. It further states that hotspots have been identified where deposition rates exceed the nearest regulatory monitor by a factor of seven (with no specific cite given). It also states that adverse ecosystem effects are directly linked to hotspots. This is a particularly strange paragraph since it is the conclusions section of the findings and conclusions chapter of the ISA yet there is no mention of N and S deposition hotspots in the balance of the ISA. There is discussion of hotspots in two ways in the ISA and its Annexes. One is in the context of hotspots of denitrification in anoxic sites in wetland soils and the second is discussion of a recent paper that used Hg levels in fish to identify biological Hg hotspots (Evers et al., 2007) which may or may not represent Hg deposition hotspots.

Based on the foregoing general and specific comments, the ISA needs extensive revision before it can form the basis for policy decisions regarding separate secondary standards for  $\text{SO}_x$  and  $\text{NO}_x$ .