



UNITED STATES ENVIRONMENTAL PROTECTION AGENCY  
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May 19, 2008

EPA-CASAC-08-011

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

Subject: Consultation 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)

Dear Administrator Johnson:

The Clean Air Scientific Advisory Committee (CASAC) NO<sub>x</sub> & SO<sub>x</sub> Secondary NAAQS Review Panel met on April 3, 2008, and has completed its consultation 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). The CASAC uses a consultation as a mechanism for individual technical experts to provide comments on the Agency's draft plan for developing technical assessments as the basis of the review of the secondary NAAQS for NO<sub>x</sub> and SO<sub>x</sub>. Written comments provided by the individual Panelists in response to the Agency's charge questions are enclosed with this letter.

As this is a consultation, we do not expect a formal response from the Agency. We thank the Agency for the opportunity to provide advice early in the NAAQS review process, and look forward to the review of the First Draft Risk and Exposure Assessment in October 2008.

Sincerely,

*/Signed/*

Armistead (Ted) Russell, Ph.D.  
Chair, CASAC NO<sub>x</sub> & SO<sub>x</sub> Secondary Review Panel

**Enclosures**

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

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## Appendix B: 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?

Appendix C: 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:**

<b>Dr. Praveen Amar</b> .....	<b>7</b>
<b>Dr. Andrzej Bytnerowicz</b> .....	<b>10</b>
<b>Ms. Lauraine Chestnut</b> .....	<b>17</b>
<b>Dr. Ellis B. Cowling</b> .....	<b>19</b>
<b>Dr. Douglas Crawford-Brown</b> .....	<b>28</b>
<b>Dr. Charles T. Driscoll</b> .....	<b>31</b>
<b>Dr. Paul J. Hanson</b> .....	<b>33</b>
<b>Dr. Rudolf Husar</b> .....	<b>37</b>
<b>Dr. Dale Johnson</b> .....	<b>40</b>
<b>Dr. Donna Kenski</b> .....	<b>44</b>
<b>Dr. Naresh Kumar</b> .....	<b>46</b>
<b>Dr. Myron J. Mitchell</b> .....	<b>47</b>
<b>Mr. Richard L. Poirot</b> .....	<b>58</b>
<b>Mr. David J. Shaw</b> .....	<b>63</b>
<b>Dr. Kathleen Weathers</b> .....	<b>66</b>

## 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?

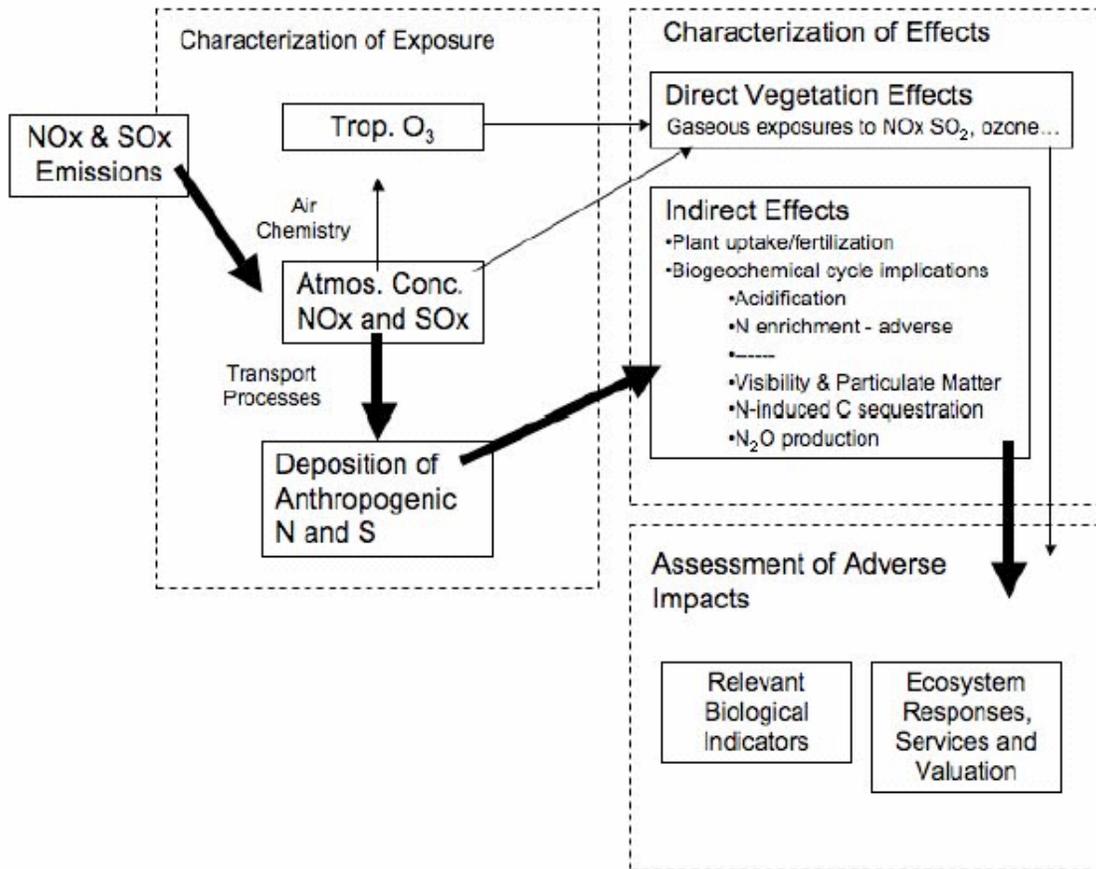
Page 2 (middle): The regulatory authority for addressing NO<sub>x</sub> and SO<sub>x</sub> as criteria pollutants is well described. More detail on how this authority translates to the subsequent extended discussion on N and S deposition effects (acidification, N enrichment) would be appropriate. A brief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

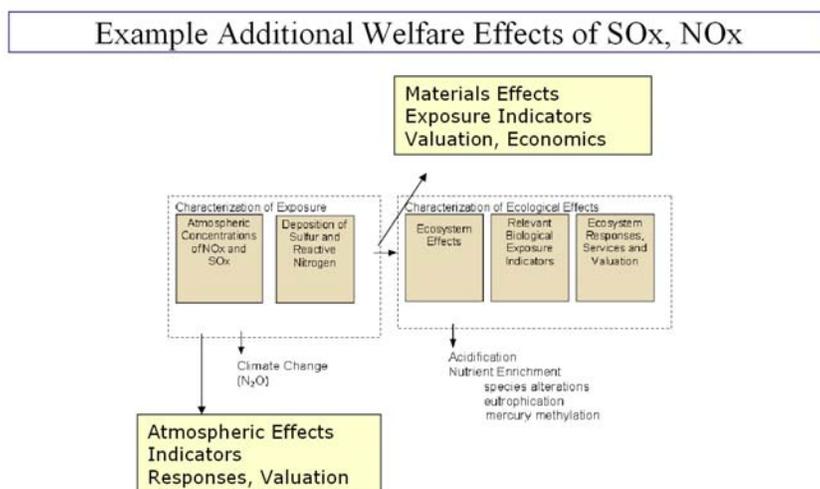
## Dr. Rudolf Husar

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

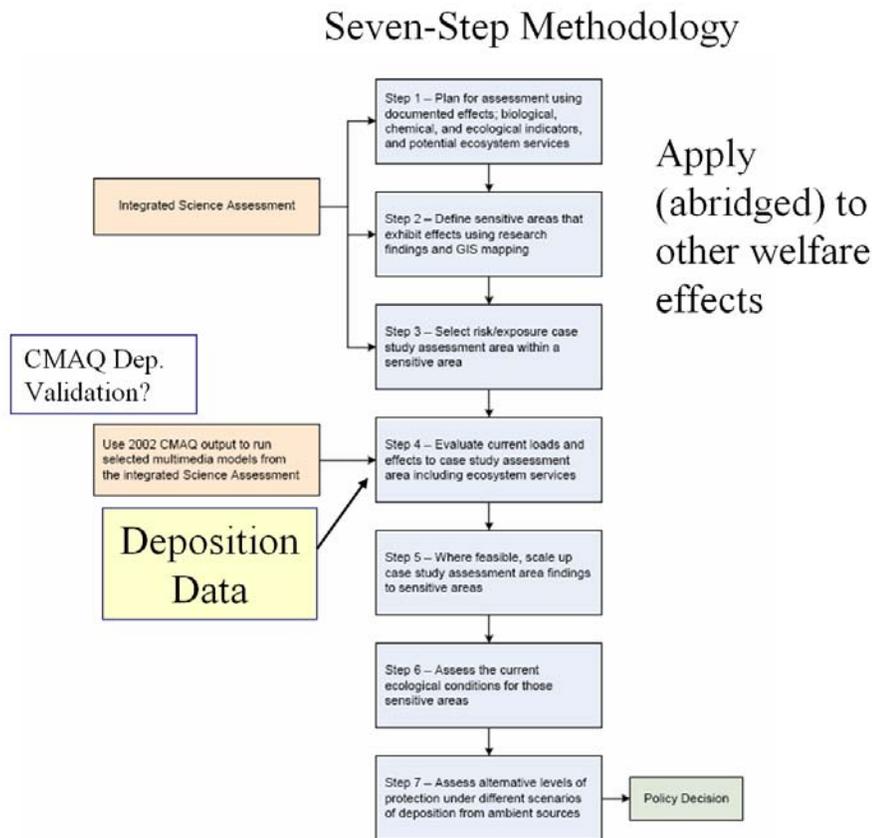
assessment. Also, as stated below, most other documents do not treat the question of indicators adequately.

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 unresolved 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.



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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 NO<sub>x</sub> and SO<sub>x</sub> 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 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.

### ***Climate Change***

Climate change discussion appears to be limited to N<sub>2</sub>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 (N<sub>2</sub>O)” slightly to “Climate Change (N<sub>2</sub>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.