30 September 2008 Preliminary Review Comments from CASAC NO_x & SO_x Secondary NAAQS Review Panel on EPA's *Integrated Science Assessment for Oxides of Nitrogen and Sulfur – Environmental Criteria (Second External Review Draft)*

Comments received:

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Dr. Praveen Amar

The specific charge is to respond to Charge Question # 3 related to Chapter 2 of the draft ISA. Specifically, the Question # 3 is noted below:

Question # 3 : Chapters 2 and 3 from the first draft have been combined. Substantially more information has been included on NH3 emissions, NH3 measurement techniques, NH3 and NH4 concentrations. Additionally, information on NOx and SOx including ambient concentrations, deposition levels, and their spatial and temporal relationships has been added. *Have these revisions to Chapter 2 improved its assessment of the currently available scientific knowledge of atmospheric sciences and its relevance to the evaluation of environmental effects presented in the later chapters*?

Response:

I would like to note that the previous Chapters 2 and 3, now combined in current Chapter 2, is a great improvement over the first ISA Draft. It is quite clear that very useful and relevant information has been included on reduced form of nitrogen (ammonia emissions, measurement techniques to estimate ambient levels of ammonia and inter comparison of these techniques, as well as concentrations of nitrate in particulate form). Equally useful is the more clear presentation on NOx and SOx (concentrations, deposition levels, trends in these levels, as well as spatial and temporal character of these two pollutants). EPA staff needs to be commended for this.

I have several specific comments that are outlined below:

1. I think the Title of the Chapter needs to change from "Source to Dose" to "Source and Exposure," or something similar. This is because this Chapter is NOT about dose. In fact, a quick "search" shows that the word "dose" appears only once in this Chapter of more than 200 pages (page 2-116 to be exact). This is not surprising because this Chapter talks about emissions, concentrations, measurement techniques, trends, etc. but dose not talk about dose. In fact, Chapter 3 is about "dose-response" and the word 'dose" appears in that Chapter frequently and correctly.

2. On page 2-2, please describe N cascade of Galloway in a little more detail since this is an important concept and not widely understood.

3. Page 2-19: I do not understand "compensation point" of ammonia. Needs some clarification. What is being compensated? Ammonia?

4. Page 2-20: Line 10: I do not think "biogenic production" of ammonia from agriculture (chiefly from livestock) is the right word. It is as anthropogenic as it can get!

5. P (O3): Ozone *Production* Efficiency: It is an important concept. However, I do not think it is clearly defined in the document (as number of molecules of ozone produced per molecule of NOx over a certain time period and over a spatial extent, etc.). I suggest it should be explicitly defined first time it appears. Just like N cascade, it is important concept and needs to be explained clearly.

6. Page 2-20: Para under the Figure: I had made this comment on the first ISA Draft. Ammonia emission totals are dominated by livestock/fertilizer operations because they simply are. They are dominant source of ammonia in their own right and not because mobile sources are not. Please drop the words "for these reasons…" on line 9.

7. I have not thoroughly read the section on Halogen Chemistry (page 2-41). Please make sure it is improvement on the version included in first draft and that it is integrated with the general theme of its relationship with, and effect on, SOx/NOx/NH3 chemistry.

8. A general comment: please be consistent and clear that you mean to give quantitative values of nitrogen and sulfur deposition in the units of "N" and "S" for kg of N/ha/yr, and kg of S/ha/yr., etc. If they are given in terms of sulfate (SO4) or nitrate (NO3), they should be clearly labeled as such. I recommend "N" and "S".

Dr. Andrzej Bytnerowicz

Question 4. Have the revisions improved the characterization of the ecological effects?

In general, Chapter 3 present a comprehensive information on ecological effects of SOx and NOx. The chapter is logically organized, contains current knowledge supported by large number of informative tables and figures. Good examples of very informative tables are Table 3-12 presenting summary of knowledge on the effects of fire on nutrient concentrations in forests in California and Nevada and Table 3-15 summarizing N effects on forest carbon cycling. In summary – the revisions improved characterization of the ecological effects in the document.

Summaries in the end of each sub-sections are very useful. Remark - summaries from Section 3.2 are not listed in the Table of Contents.

References in the text and list of citations are not always matching. Some of them are not cited properly. Examples of this deficiency are the references of Bytnerowicz et al., or Temple and Taylor (1983), incorrectly cited in the text as Temple (1983).

Remark - careful technical editing of a final version of this document is recommended.

Question 4a. Chemical effects of acidification in aquatic ecosystems

Information on acidifying effects of NOx and SOx aquatic ecosystems is comprehensive. Conceptual diagrams for this section have been correctly chosen. Examples of such diagrams are Figure 3.1 presenting fluxes of major ions associated with S depositions or Figure 3-5 showing the effects of acidic deposition on Ca supply in trees, their physiology, growth and resistance to environmental stresses.

Discussion of interactions between acidification and plant disease is well written and up to date. Interaction of acidity with foliar, membrane-associated calcium, which influences responses of trees to changing environmental conditions, such as cold temperatures and winter injury, is very well described. Remark - a reference to Figure 3-5 should be made in the text.

Question 4b. Quantitative relationships between nitrogen deposition and ecological effects

Information on the published critical loads data from Europe is mostly contained in Table 3-24 adapted from Acherman and Bobbink (2003). The table summarizes biological indicators and related critical loads for major ecosystem types in Europe and is probably sufficient for this document. However, I would like to recommend that the recent reports of the ICP Forests and ICP Modeling and Maps are also considered for an updated information on CL modeling efforts in the European forests. Summary of the dose-response curves for N deposition and ecological indicators presented in Table 3-25 is valuable and should greatly help in developing a large-scale evaluation of critic loads in the U.S and its spatial extrapolation.

The nitrogen enrichment section has been expanded and contains discussion on carbon budgeting, and fluxes of biogenic nitrous oxide and methane. Section on the carbon budget as related to N deposition is comprehensive and based on the recent scientific literature. Figure 3-35 shows interactions between the N and C cycles while Table 3-15 summarizes N effects on forest carbon cycling in Europe and North America. The section is well organized and written and is supported by informative graphs and summary tables. Minor remark – page 3-136, citation of Sutton et al (2008) does not seem to be correct. Section on methane and nitrous oxide flux is based on the meta-analysis of the recently published studies. The presented evidence shows that N deposition results in higher concentrations of these greenhouse gases in the atmosphere.

Question 4c. Other welfare effects

Good discussion of direct effects of nitric acid is provided . However, it should be emphasized that the described damaging effects of HNO3 have been demonstrated in controlled experiments for relatively short-term exposures at highly elevated concentrations of the pollutant. Long-term effects of lower, closer to ambient, levels of HNO3, should be investigated. Similarly, visible injury to plant foliage due to NO, NO2 or NH3 exposures have been demonstrated only at very high concentrations (controlled studies, industrial spills), at the levels that normally do not occur in ambient air.

Table 3-28 on direct effects of SO2 should be moved closer to the text describing these effects.

Remark - please see my above comments regarding a need for correct citation of references in this sub-section.

Executive Summary

Although it generally adequately summarizes the ISA document, there are several deficiencies that should be resolved:

1. In the Introduction, or in a footnote to it, it should be noted that 1 ppm = 1000 ppb. Readers less familiar with science may not know this fact, and both units are used throughout the text.

2. In the section "Current concentrations and deposition in the US", there are some statements that are nor correct. For instance, on page 2, last paragraph of the first column, a statement is made that the highest mean N deposition of about 9 kg/ha/yr totals is in the Ohio River valley. This value seems to be too low – I suspect that this is only the wet portion of N deposition. On the same page, the second paragraph of the second column, states that the model-predicted values in some regions of the Adirondacks are >20 kg N/ha/yr. In southern California the model-predicted total N deposition can be as high as 32 kg N/ha/yr. Actually, according to Fenn (2008), the highest values can be as high as >100 kg N/ha/yr on the western slopes of the San Bernardino Mountains. However, that high deposition mostly results from deposition of HNO_3, NH_3 and particulate NO_3^- and NH_4^+, not NO and NO_2 as the Executive Summary states.

3. In the section "Ecological effects of N deposition" a clear statement is needed that ecological effects of NO_x and SO_x should be considered in a context of often unpredictable climate changes. Ecological effects of NO_x and SO_x will also depend on other stressors, such as elevated background levels of ambient ozone, drought, insect and pathogens outbreaks, etc.

4. on Page 6, 3rd paragraph of the first column – a statement "Exposures of NO_2, nitric oxide (NO), peroxyacetyl nitrate (PAN) and nitric acid (HNO_3) cause similar forms of plant injury and decreased growth" is not true. Characteristics of foliar injury caused by these various compounds are actually quite different (please see Bytnerowicz et al., 1998 for discussion of this issue).

Ms. Lauraine Chestnut

Charge Question 1: Executive summary and key findings

The Executive Summary (ES) is a useful addition to the document, but it needs a little more detail to communicate more effectively the nature and magnitude of current harmful effects on ecosystems from N and S deposition in the US. As currently written, it describes the effects that can happen but says too little about the extent to which these are currently happening.

The "Scope" section needs a paragraph or two explaining the transport of emissions and their interaction in the atmosphere that result in N and S deposition. The first paragraph in Section 4-1 on page 4-1 in the conclusions section provides a description of this at a level of detail that would be useful in the ES.

A paragraph or two on the major sources of emissions that lead to N and S deposition would be helpful in this section. This information is well described in Chapter 2.
The "Current concentrations" section needs to reconcile the information about declines in ambient concentrations from 1990 to 2005 versus the "10-fold increase" in deposition in the past 100 years. It would be appropriate here and in the Introduction chapter to mention the Acid Rain Program (Title IV) as one of the main reasons for the decrease in emissions (especially SO2) from 1990 to 2005 and to note that the reductions from this program are now close to fully implemented and are leveling off. This program is not part of the NAAQS process, but it seems odd not to mention such a major program that was motivated by concern about the effects of acid deposition.

In the section on "Ecological effects of acidification" there needs to be more descriptive information about the effects and their current extent and significance in the US. I'm not sure it comes through clearly that certain species of fish, particularly ones that people like to catch, have died off and cannot live in these acidified lakes and streams. I'm not sure the words "decline in fish species richness" quite gets the significance across. The discussions of the Adirondacks and Shenandoah and expectations of future acidification conditions at current deposition levels are good and are examples of the kinds of descriptive implications appropriate for the ES.

In the section on "Ecological effects of N deposition" there needs to be more descriptive information about the extent and significance of eutrophication of estuarine ecosystems in the US. How many of the major estuaries in the US show significant eutrophication?

What is the extent of losses to fisheries and habitat?

The section on ecosystem services affected by N deposition (section 3.3.9) is good. It would be good if something similar were done for acidification in section 3.2.
Charge Question 2: Chapter 1
Some discussion of how Title IV (Acid Rain Program) fits into this history would be appropriate. Also some mention of the 1995 "Acid deposition standard feasibility study" report to Congress would be appropriate as part of the background.

Dr. Ellis Cowling

General Comments on the New NAAQS Review Processes

Before dealing with the details of my specific assignments during the October 1-2, 2008 meeting of CASAC I would like to offer a few general comments about the **current efforts to streamline the NAAQS review processes** and **how these changes in process are playing out in the case of the CASAC Review of the Secondary (public-welfare based) NAAQS for NOx and SOx.**

Many of us in CASAC have been very pleased that EPA has recently shown increased willingness to think more holistically – and in more fully integrated ways – about both the policy-relevant science and the practical arts of air quality management aimed at protection of both public welfare and public health. These shifts in both emphasis and approach have included:

1) Increased emphasis on scientific questions that are as directly relevant as possible to well-defined policy questions of concern to EPA. This shift to greater policy relevancy in our scientific discussions within CASAC will increase efficiency in the preparation and CASAC review of NAAQS documents, and also help streamline the NAAQS review Process. For example, the following policy-defining statement was recommended as a guide during NAAQS review processes:

   "What scientific evidence and/or scientific insights have been developed since the last review to indicate if the current public-health based and/or the current public-welfare based NAAQS need to be revised or if alternative 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?"

2) More frequent discussion about both public-welfare and public-health impacts of mixtures of air pollutants;

3) Separation of the preparation and review of documentation for a Secondary (public-welfare-based) NAAQS from the (previously always dominating) Primary (public-health-based) NAAQS review processes,

4) The decision by the Science Advisory Board (SAB) of EPA in January 2007 to establish a special Integrated Nitrogen Committee (INC) with the following set of objectives:
   a) Identify and analyze, from a scientific perspective, the problems nitrogen presents in the environment and the links among them,
   b) Evaluate the contribution an integrated nitrogen management strategy could make to environmental protection,
   c) Identify additional risk management options for EPA's consideration, and
   d) Make recommendations to EPA concerning improvements in nitrogen research to support risk reduction,

5) The unprecedented decisions by EPA to undertake an integrated [simultaneous] review of the Secondary NAAQS for two Criteria Pollutants at the same time [Oxides of Nitrogen (NOx) and Oxides of Sulfur (SOx)], and to facilitate the required CASAC and public reviews of:
    i) an Integrated Science Assessment (ISA) for NOx and SOx -- to be issued in December 2008,
    ii) a Risk/Exposure Assessment (R/EA) -- to be completed by July 2009,
    iii) a Policy Assessment/Rulemaking document prepared in the form of an Advanced Notice of Proposed Rule Making (ANPR) -- by August 2009, and thus to achieve
    iv) Final Rule Making -- prior to a court-ordered deadline of October 19, 2010.

In view of this demanding series of deadlines following our October 1-2 CASAC meeting, it is obvious that EPA and CASAC now have only about:

i) **3 months to complete the Final ISA for NOx and SOx**, and
ii) **11 months in which to complete reviews of both the First and Second External Review Drafts and then to prepare a Final Draft R/EA for NOx and SOx**.

But we now have the considerable advantage that the new NAAQS review process envisions preparation of **much more concise and much more policy-focused ISA and R/EA documents** for the present Integrated Secondary NAAQS for NOx and SOx than has historically been achieved in the **encyclopedic Criteria Documents** that have been prepared during the years since 1970.

Specific Comments on the ISA for the Secondary Nitrogen and Sulfur Pollution NAAQS Standards

The present Second Draft ISA is a very huge volume which consists of:

Chapter 1 – an appropriately short (10 page) "**Introduction**,"

Chapter 2 – an exhaustive (202 page) analysis of "**Source to Dose"** relationships for both NOx and SOx,,

Chapter 3 – a similarly exhaustive (237 page) analysis of **Ecological and Other Welfare Effects**. This chapter is appropriately focused on five critically important impacts of atmospherically deposited reactive nitrogen and sulfur compounds – acidification and nutrient enrichment of aquatic and terrestrial ecosystems and sulfur induced increase in methylation of mercury,

Chapter 4 – a reasonably short (33 page) **Summary and Conclusions** chapter that includes many statements of conclusion from the science that is reviewed Chapters 2 and 3,

Glossary – a very neat set of definitions for many of the specialized terms used in Chapters 1-4,

References – an exhaustive (124 page) list of (nearly 3000 scientific literature citations!) most of which are relevant to the possible revision of the current NAAQS standards for nitrogen and sulfur pollution. It would be very valuable to identify a limited number of these documents that are especially significant in terms of the policy purpose for which this ISA was written.

Having carefully reviewed all of these several parts of this Second Draft ISA, I was:

1) **Generally satisfied with the parts of Chapters 1-3 that deal with oxides of nitrogen and sulfur, but was disappointed that much less attention continues to be given in Chapters 2 and 3 to the quantitative importance and significant biological impacts of chemically reduced forms of total reactive nitrogen. This is true with regard to both acidification and nutrient enrichment of aquatic and terrestrial ecosystems.**

2) **In spite of the 'relatively less attention' comment mentioned in comment 1 (above), I was very pleased to read in Chapter 4 the 23 bold-face-type statements regarding the "sufficiency of evidence to infer a causal relationship between 'acidifying deposition' and/or 'Nr deposition'" and each of the many adverse acidification and nutrient enrichment effects of atmospheric deposition reactive nitrogen and sulfur pollution on aquatic and terrestrial ecosystems that are discussed in Chapters 2 and 3.**

3) **At the same time, however, I was disappointed that almost all of the non-bold-faced-type conclusions among the Summary and Conclusions statements in Chapter 4 were notably lacking in specific articulation of the quantitative importance and significant biological impacts of chemically reduced and organic forms of reactive nitrogen in both terrestrial and aquatic ecosystems.**

4) **This lack of specific articulation of the quantitative importance and significant biological impacts of chemically reduced and organic forms of reactive nitrogen was especially worrisome in the final "Conclusion" statement in the Executive Summary of this ISA.**

<u>Specific Comments on the Executive Summary of the ISA</u>

This final Conclusion in the Executive Summary includes the following two statements both of which are incorrect and misleading in terms of their relevance to the policy purpose for which this ISA is being written:

> **"Acidification is driven by the deposition resulting from NOx and SOx pollution."**
>
> **"In addition to acidification, deposition resulting from NOx, along with other sources of reactive nitrogen (e.g., fertilizers, waste water, and atmospheric ammonia deposition), causes a suite of ecological problems including biodiversity losses, disease, eutrophication, and harmful algal blooms."**

Permit me to offer the following improved and more accurate wording of the final Conclusion of the Executive Summary (please note especially the explicit recognition of both reduced and oxidized forms of reactive nitrogen -- both of these chemical forms of nitrogen are significant "Pollutants of Concern" as well as causal factors in both acidification and nutrient enrichment:

Conclusion
The three main effects of nitrogen and sulfur pollution presented in this ISA are acidification, nitrogen enrichment, and methylation of mercury. Acidification of ecosystems is driven primarily by atmospheric deposition of NOx, **NHx,** and SOx. These three pollutants cause a cascade of effects that harm both aquatic and terrestrial ecosystems including localized extinction of fish populations and other aquatic species, and slower growth and injury to forests. In addition to acidification, atmospheric deposition resulting from NOx **and NHx** emissions, along with other sources of reactive nitrogen (e.g., fertilizers and waste waters) cause a suite of ecological problems including biodiversity losses, disease in aquatic and terrestrial organisms, as well as eutrophication and harmful algal blooms in both fresh water, estuarine, and ocean ecosystems. Both gaseous sulfur compounds and particulate sulfate can interact with methanogenic bacteria to produce methyl mercury, a powerful toxin that can bioaccumulate to toxic amounts in food webs at higher trophic levels (e.g., otters, king fishers.)

Specific Comments on the Value of the Recently-Posted Document Titled *"Selected Recommendations and Findings from the Integrated Nitrogen Committee"* and an earlier *Resolution by* the Integrated Nitrogen Committee of EPA's Science Advisory Board

As many of us on the CASAC NOx/SOx Secondary NAAQS Panel will recall, during our October 30, 2007 CASAC meeting, the following *Resolution* was received from the SAB's Integrated Nitrogen Committee.

Resolution from the Integrated Nitrogen Committee of the Science Advisory Board -- for Consideration by the CASAC Secondary NAAQS NOx and SOx 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 NOx and SOx during our CASAC Conference Call Consultation on October 30, 2007.

Resolution

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

This Resolution is an important part of the scientific foundation and rationale behind my concern that the "Second External Review Draft of the ISA for Oxides of Nitrogen and Sulfur – Environmental Criteria" and the First Draft of the "Risk and Exposure Assessment for Review of

the Secondary National Ambient Air Quality Standards for Oxides of Nitrogen and Oxides of Sulfur" are not consistent with the central idea of this Resolution – that both chemically reduced and chemically oxidized forms of reactive nitrogen must be considered in the current round of reviews of the NOx/SOx Secondary NAAQS Standards.

Further justification for these ideas -- and policy recommendations that derive from the extended deliberations of the SAB's Integrated Nitrogen Committee -- are contained in a recently completed Summary Document titled: "*Selected Recommendations and Findings from the Integrated Nitrogen Committee*." This document was recently posted on the SAB website and thus is available for public review and study by any who are interested. Dr. Ted Russell, Chair of our CASAC Panel for review of the NAAQS for NOx and SOx Secondary Standards has recommended that I include a copy of this document in my individual comments so it can be considered by all members of our NOx/SOx NAAQS review Panel. An electronic copy of this same document (including properly formatted copies of the two figures and single table) is also accessible at the following URL: provided by Kyndall Barry – the DFO for our CASAC Panel:

http://yosemite.epa.gov/sab/sabproduct.nsf/0/EFB3E4663E2143F885257465006C5B32?OpenDocument&Date=10/20/2008

Dr. Paul J. Hanson

I found the second draft of the Integrated Science Assessment (ISA) to be greatly improved. Panel suggestions on the first draft appear to have been incorporated for the most part. The requested Executive Summary is well crafted and a nice introduction to the ISA, but some wording changes may still be in order. My specific comments and editorial suggestions are listed below.

Specific comments and suggested edits:

Executive Summary

Page two, second column: Change to "Numerical modeling experiments can help fill in these data gaps and suggest that local and even regional areas of high concentration and deposition may exist *where measured data are unavailable*. Subscript the 2 in NO_2.

Page 4 last paragraph: The last statement is not correct. The onset of leaching is not a fixed constant that applies to all eastern forests. This text should be changed to indicate that such a threshold applies to a defined set of sensitive eastern forests.

Page 5: Change the first two sentences to: "N deposition often increases primary productivity*, but may not lead to enhanced carbon sequestration*." In the first paragraph change "However, alteration…" to 'Alteration…' I would add the word possible to the following: "The increase in growth is greater for some species than others, leading to *possible* shifts in population dynamics, species composition, community structure and, in extreme instances, ecosystem type." Growth changes are not a guarantee of cascading effects. The paragraph on lichens seemed too general. Are the limits for lichen response true for all ecosystems, or only sensitive ones?

Page 5 last paragraph: Should organisms be animals? Elemental mercury is taken up by plants.

Page 6 third paragraph: Change to "*Acute exposures* to NO_2,…." Delete the word "Overall".

Page 6 last paragraph: Consider the following changes:

> "…deposition resulting from NO_X and SO_X pollution. It causes a cascade of effects that harm ***susceptible*** terrestrial and aquatic ecosystems, including slower growth and injury to forests, and localized extinction of fishes and other aquatic species. In addition to acidification, deposition resulting from NOx, along with other sources of reactive nitrogen (e.g., fertilizers, wastewater, and atmospheric ammonia deposition), causes a suite of ecological ***changes within sensitive ecosystems*** including biodiversity losses, disease, eutrophication, and harmful algal blooms. Particulate sulfate can interact with methanogenic bacteria to produce methylmercury, a powerful toxin that can bioaccumulate to toxic amounts in higher trophic levels (e.g. otters and kingfishers).

Chapter 1: No comments.
Chapter 2

The font size for the text within figures on pages 2-96 through 2-100 is too small. These figures need to be adjusted.

Figures 2-41 , 2,42, 2-43, and 2-44: Using the same color scheme for an alternate range of concentrations in the comparison of Total vs. background concentrations masks what should be the main message of these figures. Background levels are very, very low. The need to visualize where background levels are highest over a range from very-low to low seems unnecessary. If the authors really want this specificity they should choose an alternate color scheme for the background map.

Chapter 4

For the bolded conclusion statements on pages 4-17 through 4-31 determine if you can change "alteration" to a statement of directional change if the data warrant (e.g., change alteration to increase in the statement of N deposition effects on N_2O emissions).

Dr. Dale W. Johnson

This draft is an improvement in some ways but I believe that it remains biased against showing any possibility of beneficial effects of N deposition and I continue to believe that this is a grave mistake. It now contains detailed descriptions of N and S cycles and recognized the fact that nitrogen (but not sulfur) is often a limiting nutrient. It now considers the potential for N deposition to enhance production and C sequestration specifically within the body of the text – and then summarily dismisses such a possibility and mentions nothing of it in summary statements. The authors clearly do not accept the possibility that N deposition can have any beneficial effects and I cannot force them to do so – I am a reviewer, not a censor. However, I will continue to voice my disagreement when asked for a review.

Curiously, it appears that the treatment of the potential benefits of sulfur are discussed much more extensively than those for nitrogen (although I fully recognize that N deposition is probably not important for crop systems, given how much they are fertilized).

Specific comments:

p. 3-7, lines 1-3: In addition to these studies, are the very detailed studies of Richter and Markewitz (2001) which show long-term soil acidification due to both tree uptake and atmospheric deposition. The fourth resampling of Walker Branch Watershed will also soon be published (Johnson et al., 2008) and I will send a copy. It documents continued declines in exchangeable Ca2+ and Mg2+ with the exception of cases where decomposing logs enrich Ca2+. The Ca declines are attributed mostly to uptake where the Mg2+ declines attributed mostly to leaching, augmented by acid deposition.

p. 3-12: I do not think the study by Bailey et al is by any means the most thorough resampling study in the US. The Richer and Markewitz study is much more comprehensive and detailed, and fully deals with the causes of soil change in a quantitative manner, which the Bailey study does not. As noted in my last review, while Bailey did evaluate the role of uptake in causing the soil changes they observed, they have grossly overestimated the potential role of acidic deposition in the soil changes they observed – it would have taken a prolonged S deposition rate of something like 200 kg ha-1 yr-1 for many decades, for example, to produce such changes as they saw in some cases. It is simply not logical to blame acidic deposition for that magnitude of change. I wrote a letter to the editor on this (Johnson, D.W. 2006. Comments on "Thirty years of change in forest soils of the Allegheny Plateau, Pennsylvania." Soil Sci. Soc. Amer. J. 69: 2077.), to which they responded, which the authors of this document should look at if they intend to highlight the Bailey studies as hallmark studies showing soil acidification by acid deposition. I noted this in my last review of this document, but this comment was apparently overlooked. The authors may choose to dismiss my letter and accept Bailey's conclusions, but they should at least acknowledge the controversy.

p. 3-13, line 33 to p. 3-14, line 1: Once acidified, it is unlikely that Al levels in soils will decline again unless the soils are limed.

p. 3-26, lines 6-12: What about the effects of N on N-deficient sytems?

p. 3-43, line 5: The base cation decline could be explained as a simple consequence of charge balance.

p. 3-75, lines 1-17: This is amazing. So we do not even know if our forests are growing at a faster or a slower rate. A fundamental piece of knowledge that is missing while we speculate about all these negative effects.

p. 3-104, lines 17-23: The beginning of this paragraph acknowledges that N can be both beneficial and detrimental – so I would add to the end of it something like the following: " or on the other hand, improved forest health by alleviation of N deficiency, increased productivity and C sequestration".

p. 3-115: Good segment on disturbance – there are also many other references on the effects of fire, including effects on water quality – see references below.

p. 3-131, lines 1-11: It is very common in commercial fertilizer studies as well as pollution N addition studies for trees to take up only a fraction of applied N unless it is applied to foliage. Even so, as noted on lines 9-11, growth increases to this uptake are common.

p. 3-133, lines 1-8: This paragraph, in my view, is biased. Of course when you add a limiting nutrient you will run up against the next limiting nutrient – this is very well know. Is this then a wholly bad thing or was the addition of the limiting nutrient "good" to start with? This seems like a blind focus on the negative to me. Why is that.

p. 3-133, lines 9-12 through p. 3-133, lines 1-10: Again, I see unnecessary bias here. By my reading, there were 6 positive responses to N listed in Table 3-15, and some were at the "moderate to high" levels of N addition. I count three negative responses, including the one by McNulty which is so prominently highlighted, and three which showed both positive and negative responses, depending on time and which species was being looked at. AND, I will wager that if you included fertilization studies in commercial, fast-growing forests, you would find a very high proportion of positive growth responses indeed.

p. 3-135, Regional Trends…. It is a shame that we do not have the data to know whether forest growth has increased, decreased, or stayed the same from the forest inventory system. The Europeans have this, yet we seem not to. That being the case, apparently, I do not find this segment particularly illuminating, although it may the best we can do.

p. 3-136, lines 3-17: I am glad that the authors included a discussion of the Magnini paper – even though they do dismiss it rather easily. I am not sure I agree with that – nitrogen is, as is acknowledged in this document – a limiting nutrient and it seems highly probable that adding it will cause increased growth. I do not understand why there is such resistance to this concept, but the authors opinions obviously differ from mine. At least it was discussed.

p. 3-162, Trees: Again, the view here is that there is nothing good about "altered growth rates". Can't altered growth rates be a "good" thing if they are in the positive direction and in, for example, commercial forests? Does this not bear even the slightest mention?

3-163 to 3-175: A very good review of N effects from the pollutant point of view. Do traditional forest fertilization studies tell us anything more?

3-164 Grasslands: It would be good to mention the cheatgrass issue in the Great Basin here. Cheatgrass is a nitrophile and increased N deposition will undoubtedly facilitate the spread of this noxious species also.

p. 3-191, line 13: I would add "increased growth" after "sensitive"

p. 3-206, lines 4 and 8-18: "forest yields" is mention in the headlines, but no mention of increased timber yield is mentioned in the following paragraph – all is negative. Why is this.

p. 3-212 to 3-218: A very good, thorough and objective treatment of sulfur.

General References

Johnson, D.W., D.E. Todd, C.F. Trettin, and P.J. Mulholland. 2008. Changes in Soil Exchangeable K^+, Ca^{2+}, and Mg^{2+}in Forests of Walker Branch Watershed, 1972-2004. Soil Sci. Soc. Amer. J. (in press)

Richter, D.D., and D. Markewitz. 2001. Understanding Soil Change: Soil Sustainability over Millennia, Centuries, and Decades. Cambridge University Press. 255p

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Dr. Myron J. Mitchell

as of Sept. 28, 2008 (Incomplete)

General Comments

Be consistent in the order of multiple citations throughout the document. Either cite by date or alpabetically. Within the document the citations are not complete or consistent including the need to use letters for multliple citations by the same author and years.

EPA Questions to be addressed:

1. We have added an executive summary of the major findings and conclusions to the second draft ISA. We have also created a "key findings" section that is intended to provide highlights of these conclusions. We are seeking CASAC panel advice and comments on these additions to the ISA. To what extent do they provide an appropriate level of detail and convey the important scientific conclusions of the assessment?

In general the information level in the executive summary is suitable. Some changes are suggested within my detailed comments.

2. Chapter 1 has been revised to clarify the scope or focus of this assessment on effects related to the deposition of nitrogen and sulfur compounds. In addition, we have added a discussion of the framework for evaluation of causality for assessing ecological effects. Do these revisions adequately characterize the scope of the assessment? Does the CASAC panel have recommendations for revisions to the causality framework? Is it appropriately applied in the draft ISA?

The information provided in Chapter 1 seems appropriate with respect to the overall approach.

3. Chapters 2 and 3 from the first draft have been combined. Substantially more information has been included on NH_3 emissions, NH_3 measurement techniques, NH_3 and ~+ concentrations. Additionally, information on NO_x and SO_x including ambient concentrations, deposition levels and their spatial and temporal relationships has been added. Have these revisions to Chapter 2 improved its assessment of the currently available scientific knowledge on atmospheric sciences and its relevance to the evaluation of environmental effects presented in later chapters?

There is certainly more detail on atmospheric chemistry in Chapter 2. I am not certain whether all of this detail including issues related to analytical technique, artifacts, differences in instrumentation, etc. is necessary with respect to having sufficient information to set standards. More explicit linkages need to be made on what we know as well as what we do not know and how this information is needed to setting standards for NO_x and SO_x. The details provided in this section are somewhat overwhelming including extensive graphics and results from specific sites. A clear summary of what we know and what we don't know with a focus on the standards would

help bring all of this information into focus. In particular the issues relating to the estimates of deposition velocities need to have a central place in this document. The flow of information could be improved with better use of headings to direct the reader to the content of major sections. Also, some further discussion is warranted about how other factors such as climate change, extreme events (e.g. fire, hurricanes, ice storms), invasion of exotics, etc. can also have a major impact on ecosystem response and can have a major influence on the expression of acidic deposition impacts on ecosystems.

4.We removed or eliminated redundancy, added summary sections, added additional references and reorganized Chapter 3. Revisions to the ecological effects sections are given below. Have the revisions improved the characterization of the ecological effects?

This section has many problems in the citations and references. Some of the sections in this chapter are redundant. For example in section 3.2.1.3 there is a review of N accumulation and nitrate leaching and in section 3.3.2 there is a section on N enrichment effects on N cycling.

a. Consistent with CASAC comments, we expanded our characterization of the quantification of chemical effects of acidification in aquatic ecosystems, added new conceptual diagrams, and further discussed interactions between acidification and plant disease.

Certainly more details are provided. However, there is considerable redundancy in the document. A clearer delineation of particular areas of focus is needed.

b. We expanded the discussion of quantitative relationships between nitrogen deposition and ecological effects, including published critical loads in the U.S.. and Europe. In addition, the nitrogen enrichment section was expanded to include new discussions on carbon budgeting, biogenic nitrous oxide and methane. Information on the linkages between effects and both reduced and oxidized forms of nitrogen was emphasized, to the extent data were available.

5. In revising the ISA, we have incorporated additional information on the indicators of exposure and ecological effects, including increased emphasis on quantified relationships in the presentation of information of results in tables and summary discussions in Chapter 4. What are the views of the CASAC panel on our revisions to focus on quantitative relationships between airborne nitrogen and sulfur compounds and ecological indicators?

Specific Comments

Page	Line	Comment
xvii		Change to "*chloride* ion."
xviii		Change to "*fluoride* ion"
1		Within the body of the text "Nitrogen" should be changed to "nitrogen".
1		Throughout the text corrections need to be made for subscripts that are lacking in some cases (e.g., NO3 should be NO_3, NO2 should be NO_2, SOX should be SO_x, etc.).
1		Change to "In the years 2004-2006, S deposition in the United States."
2		Change "inferred" to "modeled."
2		Change to "Although deposition in most areas of the United States *was dominated by* wet deposition, there were some exceptions, including parts of California where N deposition was primarily dry."
2		Change to "The *sparse* coverage of monitoring sites in many areas, especially in the rural West, *results in little or no* data exist on deposition totals in a *substantial* number of potentially sensitive places."
2		Change to "Numerical modeling *efforts* can help fill-in these data gaps and *may* suggest that local and even regional areas of high concentration and deposition exist where *currently* no data exist.
4		Some of most severe acidification may also occur during the summer after periods of drought especially in those watersheds with substantial portions of wetlands. These rewetting episodes are associated with acidification due to the oxidation of previously reduced sulfides.
2-1	8-18	Isn't there some merit in changing the definition of nitrogen oxides to be the same as that used by atmospheric scientists and air quality control experts?
2-4	2	Change "Roughly" to "*Approximately*."
2-6	6	Change to "The N content in fossil fuels and chemical forms vary strongly."
2-6	7	Change "running" to "operating conditions".
2-7	35	I thought that the effect of "stack height" effect was relatively minor with respect to the overall dispersion distance of pollutants from combustion.

2-8	16	Change to "of the various products of *N* transformations."
2-8	22	Change to "synthetic and organic *N* fertilizers."
2-8	29	Change to "N metabolism in soils is strongly dependent on soil substrate concentrations *of both N and labile carbon constituents* as well as physical."
2-8	30	Change to "Where *available* N."
2-9	11-12	Change to "such as the *differences* between shortgrass and tallgrass prairie for example."
2-9	18-19	Change to "Local contributions to soil NO_x can be greater *per unit area* than the global average."
2-10	28	Delete extra "," in "(e.g.,,." Note this occurs twice in this line.
2-11	14-16	This statement is a little confusing. It suggests there a combination of both atmospheric residency time as well as heat trapping efficiency that produces this value of 300? These two parameters need to be clearly separated. Isn't the heat trapping capacity on a per molecule 310 times that of carbon dioxide?
2-11-12		Reword as follows: However, N_2O is an intermediate product along with NO from the complex soil metabolism described in Section 2.2.2.1. A brief description of N_2O emissions and its contribution to the U.S. GHGs is described below.
2-12	16-17	Delete: These emissions resulted from the fuel combustion, industrial practices, and stimulation of biogenic sources through agricultural practices enumerated above.
2-12	18-19	Change to "*From* 1990 and 1998."
2-12	23	Change to "Biogenic production of N_2O *from soil* accounted for > 75%."
2-12	27	Change to "may be *affected* by environmental conditions."
2-12	28-29	Change to "*enhancing* denitrification, and potentially increasing N_2O emissions."
2-13	8	Change to "emissions originate*d* from."
2-13	10	Change to "either *as* SO_2 or SO_3."
2-13	14	Change to "can not accurately be *used to calculate* the contribution of local sources to selected environmental."

2-13	18-19	Delete "with most counties east of the Mississippi River in warmer colors (greater emissions densities) than most counties in the West."
2-15	6	Change to "98 and 160 tons total SO_2 per square mile, respectively."
2-18	4	In addition to carbon-bonded S (amino acid) there is also inorganic sulfate.
2-22	1-3	This is a confusing sentence that needs to be reworded.
2-22	10	Change to "have been performed *for* a number."
2-22	12	Change to "it can be treated *conservatively* on these scales."
2-23	30	Change to "may have been underestimated or *were* increasing."
2-24	6	Change to "proved useful for *estimating*."
2-25	11	Change to "volatility. This pattern, however, has not appeared in previous emissions factors and inventories."
2-26	10	Change to "available in numerous *references* (Seinfeld."
2-26	11	Change to "recounted here with special *attention* to."
2-26	12-13	Change to "are schematized in Figure 2-15. NO_2, itself an oxidant, can react to form."
2-26	16-17	Change to "to HNO_3 and can contributing to the acidity of cloud, fog, and rain water."
2-27		Figure Caption 2-15 As stated previously it would be helpful to have the definitions of "NO_x" and "NO_y" be consistent with the atmospheric science literature.
2-29	2	Delete "highways."
2-29	7	Change to "Reaction 7:."
2-29	14	Delete "schematic."
2-29	17	Change to "this chapter are:."
2-30	2	Change to "NO_3:." Note I would suggest that if reactions are provided after a statement this statement should end with a ":" throughout the document.
2-32	15-16	Be consistent in the order of multiple citations throughout the document. Either cite by date or alphabetically.

2-34	4	Change to “decomposition ranges.”
2-34	6	Change to “uptake *by* vegetation.”
2-34	7	Change to “Reaction 21 *is* thermally.”
2-34	11	Change to “concentrations of their precursors.”
2-34	12	Change to “found in most environments, ranging from remote.”
2-34	14	Change to “downtown metropolitan areas, especially.”
2-35	5	Change to “because many factors important for $P(O_3)$ are omitted.”
2-35	6	Change to “VOCs *that* are mostly absent during early morning hours.”
2-35	11-12	Delete “at a monitoring site in Shenandoah National Park, VA.”
2-35	18	Change “from commercial aircraft are very similar.”
2-35	20	Delete “included in model calculations.”
2-35	27	Change to “(1998, 2001).”
2-35	32	What is implied by the use of the term “correlation patterns”? See also page 2-36, line 1.
2-36	7	Change to “2001 , 2003).”
2-37	10	Change to “Brown et al. (2006) found.”
2-38	1-2	Change to “available for reaction thus increasing $P(O_3)$.”
2-38	5	Change to “amounts. However, only.”
2-38	8-11	Change to: “It is important to recognize that *the studies of* both Schultz et al. (2000) and Singh et al. (1996) involved aircraft sampling at *high* altitude *that* can significantly under-represent sea salt aerosols. These aerosols are important contributors to total NO_3 (defined to be $HNO_3 + pNO_3$) and large fractions of NOY(Huebert, 1996).”
2-38	12	Change to “budgets based *upon their studies*.”
2-38	14	Change to “domine, 2002) that.”
2-38	19	Change to “*Hence*, HNO3 recycling.”

2-38	33	Delete "observed."
2-38	34	Change "(Reaction 18) *constitutes* a."
2-39	6	Change to "and kerosene soot, *and found* that."
2-39	16	Delete "observed.".
2-39	18	Change to "Longfellow et al. (1999)."
2-39	20	Change to "NO_3/N_2O_5 and HO_2/HO_2NO_2."
2-40	11-12	Change to "Daytime observations of HNO_2, when rapid photolysis depletes ambient concentrations to very low levels, implies large sources of photo-induced."
2-48	4	Change to "Jacobson (2002)."
2-48	12	Change to "~5 x 10^{-3} with."
2-50	6-7	Is there still a quantifiable plum "further downwind". Why are conditions even "more oxidizing than in background air"?
2-55	18	The correct abbreviation for moles is "mol" not "M".
2-56	2	Change to "throughout the Earth's boundary layer."
2-56	8-9	Does the term "reservoir species" imply chemical species with larger "τ"?
2-56	19-20	Change to "Photochemical activity is enhanced by higher temperatures and sunlight."
2-57	1	Change to "distances from their *sources*."
2-59	26-27	The format here is mixed up and needs a ")" at the end.
2-60	1	Change to "(2007)". There are two 2007 references that need to be designated with letters.
2-60	1	Delete "parts per billion" and "[]".
2-60	32-34	Change to "The daily average interference for an episode during the summer of 2002 was modeled. Concentration fields for NO_z species and conversion efficiencies for NO_z species ranged from ~20% in Baltimore to ~80% in Madison, VA.

2-61	1	Change to “activity *was* highest.”
2-61	2	Change to “compounds *were* greatest”
2-61	7	Delete “On the whole.”
2-61	10	What is meant by "can be changing rapidly"? Do you mean changing rapidly in the literature
2-65	7	Change to “normal” to “typical.”
2-65	12-17	These statements are important in relating the results to actual needs and determination of standards. Similar types of statements are needed throughout the document to focus the results on those issues which are policy relevant.
2-66	4-5	This sentence needs to be reworded since “and of the particles against deposition” does not make sense. See also line 9.
2-66	11-13	Delete the term “standard” in these statements. There is no actual standard configuration of these devices.
2-69	1	Clarify what is meant by “In addition to the elevation by EPA.”
2-69	2	Delete “and reported by.”
2-69	3-6	Change to “The methods included the use of a tunable diode laser (TDL) absorption spectrometer, a wet scrubbing long-path absorption photometer (LOPAP), a wet effusive diffusion denuder (WEDD), an ion mobility spectrometer (IMS), a Nitrolux laser acousto-optical absorption analyzer, and a modified CL analyzer.”
2-69	11-13	Change to: “Hence comparisons of ambient NH_3 instruments have confirmed that no single technique has yet been identified that provides automated, high quality results for continuously determining NH_3 at low concentrations.”
2-71	5	Be more explicit in using the term “positive artifact” here and elsewhere. Does this mean to suggest that there are overestimates of concentration measurements? Would a better term be something like artifacts resulting in overestimates.
2-71	11	See the comment above and modify to avoid confusion I the term “negative interference.”
2-60	7	See comment about Dunlea et al. references.

2-78-81		This section could benefit with more focus on the issues related to evaluating SO_x and NO_x at the scales important for this assessment with respect to chemical transport models CTMs.
2-81-83		Is this review of global scale issue important for the issues associated with regional/local situations. At a minimum this section could be reduced playing particular attention to those global aspects that can have regional implications.
2-87	3	In the Picketering et al. citations remove ";". These format errors need to be corrected throughout the document.
2-88		The previous pages (2-78 to 2-87) need to have a clearer linkage to this deposition section.
2-89	9-13	Certainly a major issue that needs focus are the errors and problems with estimating dry deposition and a considerable amount of these concerns related to the calculations of deposition velocities.
2-90	13-15	The statement that "Deposition rates are independent of leaf area or stomatal conductance, implying that deposition occurs to branches, soil, and the leaf cuticle as well as leaf surfaces" is misleading since leaf area and stomatal conductance are important, but that other factors also need to be included.
2-93-95		The section on "Air Quality Model Evaluation" is a critical part of this document.
2-94	3-5	These errors can lead to erroneous predictions especially with respect to those associated with future air quality estimates.
2-101-202		Figures 2-38 and 2-39 suggest that the model results are quite different than those for the NADP network sites. Doesn't this bring into question the model validity for predicting these wet deposition amounts?
2-104	12-13	Suggesting that Ext-RADM provides "good" agreement when R^2 values are between 0.4 to 0.7 for most species is misleading since at lower values less than 50% of the variation is being explained.
2-105	1-4	These statements suggest a better Ext-RADM result. Is this due to the value representing a larger spatial unit and also an estimate for a longer period?
2-108	1-4	I am not sure of the rationale for including in PRB the anthropogenic sources outside of U.S., Canada and New Mexico.
2-120		Are the measurement protocols in State and Local Air Monitoring Systems (SLAMS) networks sufficiently similar for these measurements to be comparable to CASTNET.

2-131	10	Incomplete sentence.
2-198-202		Having this summary is helpful in placing the large amount of information provided in the document in context. Some further inclusion of statements relating to problems with respect to dry deposition estimates including the issues associated with estimating deposition velocities needs to be included.
2-199	18-21	This sentence needs to reworded and likely separated into a series of sentences.
3-4	25-26	Why are wetlands given special attention here?
3-7		Figure 3-2 caption. Change to "Diagram illustrates soil horizons *commonly found*".
3-9	8	Change to "(Shanley *et al.*, 2005)."
3-9	14-15	References are not cited correctly (these are multiple authors) including a need for a letter designation for Driscoll et al. (2001)
3-9	20	See comment above regarding citation. It appears that this section has many cases where the references are not cited correctly including the absence of showing that the references are multiple authors.
3-11	9	Section "0"?
3-12	17	Another line with incorrect referencing being used.
3-12	30	Another line with incorrect referencing being used.
3-16		Figure 3-3 needs a reference.
3-17		Figure 3-4 needs a reference.
3-19	3-9	Even in the West the nitrate in surface waters during snowmelt is mostly microbially derived and is not from direct atmospheric input. These statements confuse this issue.
3-22	4	There are also large differences in tree species to tolerance of different levels of Al. Note this is indicated later in line 14. A clear statement is needed of the major factors associated with Al toxicity.
3-22	14	Figure 3-5 does not clear depict that Al stress varies with species as suggested with this reference.

3-25	2-5	The term buffering is not correctly used here. Reword to : Once base saturation decreases to a critical level (approximately 15–20%), inputs of H_2SO_4 and HNO_3 result in exchange of inorganic Al.
3-25	9-10	Change to " If the C:N ratio of soils falls below about 20 to 25, nitrification is stimulated resulting in *net nitrification and increased acidity*."
3-27		Figure 3-6 needs a reference.
3-34	18-21	Also grassland tend to be found on soils with relatively high pH and % base saturation. Aren't grassland associated with acidic soils less common?
3-39	16	Change to "in *most* affected."
3-39	22	Delete "to a greater extent than SO_4^{2-}"
3-39	23-24	Delete "The importance of NO_3^- as an agent of acidification varies by region."
3-39	29	Delete "Average."
3-40	1	Change to "Surface water NO_3^- concentrations have changed over time and these trends vary by regions".
3-40	19	Change "outbreak of gypsy moths, which consumed foliage" to "gypsy moth defoliation."
3-40	20-21	Delete " in affected watersheds."
3-40	32-33	Change to "annual air temperatures were strongly related to average annual NO_{3-} concentrations in stream water".
3-41	5-6	Was it really suggested that an increase in pH stimulated primary productivity? If this is the case this should be attributed to the authors of the article.
3-41	9	Change to "between recent *tempora*l trends."
3-42	1-2	Change to "(Aber, 2003). Moreover, spatial patterns of NO_3 concentrations in surface water across the Northeastern U.S. are consistent with atmospheric N deposition values although there is considerable variation in these concentrations based upon watershed attributes.
3-42	15-16	Change to "Within western Virginia and in Shenandoah National Park, concentrations of base cations in streams did not exhibit significant temporal trends from 1988 to 2001."
3-42	16-17	If sulfate concentrations did not change indicate this explicitly versus an indirect reference to the role of sulfate adsorption.

3-43	1-2	See comment above regarding the results from Virginia.
3-43	6-7	Delete "The pH of water quantifies the hydrogen ion concentration, which is toxic to many forms of aquatic life."
3-43	12	Explain the derivation of these three pH reference levels.
3-43	22	Change to "southwestern Adirondacks acidified more compared to other lakes in the Adirondacks since preindustrial time."
3-44	6	Should this be "(0.18 μeq/L/yr)"?
3-45	1	Be explicit in referring to this study. I assume this is Stoddard et al. (2003).
3-45	18	Do you mean by more stable that ANC is not affected by ambient CO_2 concentrations? Make this more explicit.
3-45	29	Explain these three cutoff values for ANC.
3-45	33	Change to "the oxidation of chemically reduced S-containing minerals."
3-47	11-14	Delete "ANC can be measured in the laboratory by Gran titration or calculated on the basis of the difference between the base cation sum and the mineral acid anion sum. Acidic waters are defined as those having ANC less than or equal to zero μeq/L".
3-48	33	Not sure the term "limited" is appropriate.
3-50-51		References are needed in this text to support the statements.
3-52		The following study provides additional information on ANC patterns within the Adirondacks using a mass balance approach: Ito, M. M.J. Mitchell, C.T. Driscoll and K.M. Roy. 2005. Factors affecting acid neutralizing capacity in the Adirondack region of New York: a solute mass balance approach. Environmental Science and Technology 39:4076-4081.
3-54		I believe some PnET-BGC simulations by Driscoll's group have also been done that would provide other estimates of the recovery of ANC.
3-56	8	Change to "rain *events*."
3-76	32-33	References needed to support this statement.
3-79	1-2	A major problem in making wide scale regional projections is that it is well known that the response even within relatively small areas (e.g. almost adjacent watersheds) that the response to acidic deposition can be quite marked.

3-79	3-6	It needs to be emphasized that the water pathways and soil depth can be very Important in the capacity to neutralize the effects acidic inputs.
3-79	13	It should also be stated that this is a threshold which is generally necessary, but does not always result in enhanced nitrate loss.
3-80-81		Some inclusion of issues associated with how biotic response can also be altered by other factors such as climate change, invasion of exotics, extreme events (e.g., drought, ice storms, hurricanes, etc.)
3-87		Figure 3-19. Why not cite NADP/NTN as the source versus Sullivan et al. (2006a). Also, the more recent data should be included.
3-88		In reviewing the results from the Adirondacks focus on those findings specific to the Adirondacks versus generalities associated with acidification of ecosystems.
3-89		The PIRLA I and PIRLA II projects provided some of the most definitive evidence of historical changes in acidification of the Adirondacks. Inclusion of some of the figures produced from this project would strengthen the document.
3-89-90		The inclusion of figures showing side by side comparison of MAGIC and PnET-BCG hind casting would be instructive especially if placed in the context of the PIRLA reconstructions.
3-90	8	Provide figures or tables clearly showing similarities and differences between the MAGIC and PnET-BGC simulations.
3-91		Table 3-10. Reference needed
3-102-123		The structure of the document results in redundancy with some of the information in this section also being presented in previous sections of Chapter 3.
R-30		There are two Dunlea et al. (2007) references that need to be designated “a” and “b” respectively.

Mr. Richard Poirot

These comments pertain primarily to Charge Question 2 and Chapter 1 of the 2nd draft ISA.

> **Question 2. Chapter 1 has been revised to clarify the scope or focus of this assessment on effects related to the deposition of nitrogen and sulfur compounds. In addition, we have added a discussion of the framework for evaluation of causality for assessing ecological effects. Do these revisions adequately characterize the scope of the assessment? Does the CASAC panel have recommendations for revisions to the causality framework? Is it appropriately applied in the draft ISA?**

Chapter 1 provides a clear, concise introduction to the ISA, including a brief history of past NAAQS reviews, an outline of the intended scope of the current assessment, and a proposed framework for the determination of causality in relationships between the pollutants of concern and the resulting ecological responses. Overall I think this chapter looks good, and my comments are mostly minor.

While the scope of the assessment is relatively broadly defined – to include for example consideration of ecological effects resulting from N-nutrient enrichment from deposition of both oxidized and reduced nitrogen – there still seems to be a rather intentional avoidance of questions of how such effects or environmental effects may be related to alternative kinds of NAAQS air-quality-related indicators. This question does get taken up to some extent in Chapter 8 of the Risk and Exposure Assessment. However, it might also be useful to add some discussion and display of the inter-relationships among various air quality, deposition and effects metrics as part of the ISA. It would be useful to know up front if deposition-based or critical load-type NAAQS are a possibility, or are we stuck with SO_2 and NO_2, again?

As indicated in previous comments, I think its unfortunate that the scope of this assessment was narrowed to exclude consideration of visibility effects from aerosol phase sulfur and nitrogen compounds. I think it would have been possible to copy/paste the same discussion of these effects in both the SOx/NOx and PM secondary NAAQS ISAs (and then decide which NAAQS - if any - would be most effective for addressing all the welfare effects of S & N emissions). In the current case it might also have led to consideration of useful alternative NAAQS indicators – which consideration of aerosol concentrations & effects would help justify. It can also be noted that the working definitions of NOx and SOx are somewhat awkward here, as NOx is taken to include nitric acid and aerosol nitrate, while SOx includes only the gaseous oxidized sulfur compounds.

The historical summary of the secondary SOx and NOx NAAQS reviews is exceedingly brief, and it seems incomplete for example to mention the 1984 EPA Acid Deposition Critical Assessment Document without also mentioning the 1990 NAPAP State of the Science and Technology and Integrated Assessment Reports, the 1990 CAA Amendments (Title IV), and 1995 EPA Acid Deposition Standard Feasibility Study Report to Congress. Granted, these were not part of the NAAQS review per se, but the 1996 decisions not to revise the secondary SOx or

NOx NAAQS were related to these other activities, and represented a clear decision that the environmental effects of these pollutants were better addressed by other regulatory mechanisms.

The proposed framework for evaluation of causality is logical and clearly presented. So far as I can tell, it appears to be appropriately (and effectively) applied elsewhere in the document – where chapters 3 and 4 contain a number of clear, persuasive statements that "**the evidence is sufficient to infer a causal relationship between** …X and Y". With the exception of a few such statements on direct vegetation effects from exposures to specific gaseous S and N compounds, the identified "causal agents" are almost always either "**acidifying deposition**" or "**reactive nitrogen deposition**", and thus appear to be intentionally non-specific to the traditional criteria pollutant definitions. I think this is fine – even preferable – if secondary NAAQS are being considered that might be deposition-based, combine S&N or combine oxidized and reduced nitrogen. If such alternative metrics are not being considered, than maybe some of these bullets might be rephrased in more pollutant-specific terms. Also perhaps there could be some causality conclusions in chapter 2, in which causal relationships might be inferred between (current and historical) emissions, ambient air concentrations and deposition of various S and N compounds.

Possibly also, the concept of a "significantly contributing factor" could be introduced here (and used later) as a sort of subset of a "causal factor" (i.e. "the evidence is sufficient to infer a causal relationship between Nr, for which oxidized N is a significantly contributing factor, and Z effects…"). This concept of significant contributing factor could be important in considering effects resulting from pollutant mixtures, as well as for considering effects which result from or are modified by the cumulative influences of both current and historical pollutant deposition.

I think the description of the 2-step process in the causality framework could be more clearly presented. There's a bit of a logic problem with saying first we will determine the causal relationship and then we will determine what effect has been caused. In reality, the process seems to be more one of starting with identifying rather broad, general causal relationships (for example between total Nr deposition and alteration of terrestrial species richness) and then in step 2 winnowing this down to identify more pollutant-specific and/or species-specific relationships at specific ranges of concentrations/exposures. Additional confusion is introduced by apparent inconsistencies between the 2 steps in the causality framework as described in lines 4 and 5 page 1-7, and the subsequent discussion of 2 steps in sections 1.6.1 and 1.6.2. In the first case, step 2 refers narrowly to the determination of whether levels of exposure can be defined at which effects of concern can be observed (kind of a yes/no threshold answer). In the second case (section 1.6.2), step 2 is more broadly and comprehensively defined to consider effects over a range of exposure conditions with evaluation of the shapes of exposure-response or concentration-response functions. Presumably this would also include consideration of estimated future ecological responses to changes (increases or decreases) in pollutant concentration and deposition. For the most part, this ISA seems much more focused on step 1 type conclusions – although there are some very useful tabular presentations of quantitative relationships between N deposition levels and specific ecological effects such as in Table 4-4. I wonder if the intent is to intentionally focus on step 1-type conclusions in the ISA and then develop more detailed quantitative evaluations of ecological response (i.e. step 2) in the Risk and Exposure Assessment. If this is the intent, why not make it clear?

Mr. David Shaw

General Comments

The executive summary is an excellent contribution to the ISA. This addresses some of my previous concerns regarding a clear message of the document.

Executive Summary, Page 2:
The paragraph which begins with "Expanding urbanization…" has a last sentence explaining heterogeneous deposition. This last sentence might be benefited by the addition transport.

It seems that it would serve the assessment better to have Section 1.7 (page 1-10) appear at the beginning of Chapter 1.

Charge Questions

Chapter 1 has been revised to clarify the scope or focus of this assessment on effects related to the deposition of nitrogen and sulfur compounds. In addition, we have added a discussion of the framework for evaluation of causality for assessing ecological effects. Do these revisions adequately characterize the scope of the assessment? Does the CASAC panel have recommendations for revisions to the causality framework? Is it appropriately applied in the draft ISA?

I feel that the scope has been properly modified to include both oxidized and reduced forms of nitrogen. Furthermore, I am pleased at the use of "acidifying deposition" throughout the document. This reflects the actual concern of this type of deposition.

Section 1.1, Page 1-3:
I still feel that particulate matter should not be omitted from this document. As my previous comments state:

PM plays a significant role in nitrogen and sulfur deposition. I feel that separating out the effects of gas- versus aerosol-phase S/N will be difficult, since wet and dry deposition can include both phases, and atmospheric chemistry and transport affect both phases. The ISA clearly states that "particulate NO_x and SO_x will be addressed with the secondary PM NAAQS review," and it therefore becomes crucial that these two review process tracks are highly consistent with each other. One cannot proceed independently of the other track.

Section 1.6, Table 1-1:
While the causality framework does seem appropriate, it is unclear as to how much weight is given to each aspect. I do appreciate that it clearly states that that scientific evidence will not have to meet all of the aspects of causality, but it might benefit the group to understand how each will be considered in the weighting process (i.e. if no consistency then not used).

Section 1.6.1, Table 1-2:
Again, while the descriptors for weight of evidence seem appropriate, I wonder if these could somehow be associated with the causality aspects. For example, given a "weight" in the form of a number for each aspect, add all the aspect numbers for a total. When this total is calculated, apply to the weight of evidence table. Perhaps "sufficient to infer a causal relationship" is used for aspect numbers of 30 or higher (depending on how the number scheme is set).

Misc.

Page 1-6, line 16:
Should read "…analyses used appropriately and…"

Executive Summary
There is inconsistency in how NO_x and SO_x is typed.

Selected Recommendations and Findings from the Integrated Nitrogen Committee

EPA Science Advisory Board

September 2008

Introduction

Reactive nitrogen (Nr) encompasses biologically active, chemically reactive, and radiatively active nitrogen compounds. At the global scale, human activities now create more Nr than natural terrestrial ecosystems produce or can assimilate. As a result, Nr is now accumulating in the environment.

Natural and human activities can release Nr to the environment in many different chemical forms. As it moves through the environment, Nr can cause both beneficial and adverse effects. The nitrogen cascade describes the movement of Nr through the environment and the resulting effects. Natural processes or control measures can change one form into another that may have different effects.

Some problems from excess Nr (associated with sewage, fossil fuel combustion, crop/animal production, etc.) are well recognized and addressed. EPA has taken an impact-by-impact approach to regulation Nr, which, with few exceptions, addresses specific forms of nitrogen in a single system (aquatic, atmospheric, or terrestrial). The principal regulatory authorities pertaining to nitrogen are the Clean Water Act (CWA) and the Clean Air Act (CAA). Because such approaches rarely consider more than a small part of the nitrogen system, they can merely delay larger scale and sometimes unanticipated impacts. They seldom prevent them. The deliberate integration of Nr research, management, and control strategies across media and issues can help maximize the beneficial uses of Nr, while minimizing adverse environmental impacts.

The Science Advisory Board (SAB) advises the Administrator of the Environmental Protection Agency (EPA) whose mission is to protect human health and the environment. The SAB's INC objectives are:

1. Identify and analyze, from a scientific perspective, the problems nitrogen presents in the environment and the links among them;
2. Evaluate the contribution an integrated nitrogen management strategy could make to environmental protection;
3. Identify additional risk management options for EPA's consideration; and
4. Make recommendations to EPA concerning improvements in nitrogen research to support risk reduction.

Nr Inputs to US

It is critical to understand the relationship between inputs of newly created reactive nitrogen vs. how much of the Nr is transferred to other compartments, as well as the effects excess Nr has on humans and the ecosystem if effective control strategies are to be developed. The largest sources of Nr created by human action in the USA are fossil fuel combustion and food production. The Nr that comes from fossil fuel combustion is chiefly in the form of NOx emissions into the atmosphere; this introduces about 5.5 Tg N per year into the environment (combustion of wood and other forms of biomass generally occurs at temperatures too low to convert N_2 to Nr). Food and turf production add about 10.9 Tg N per year from fertilizer use and another 7.7 Tg N per year due to cultivation-induced biological fixation. Industrial activities introduce an additional 4.2 Tg N per year into the US. Imports of commodities contribute another 0.2 Tg N per year (a teragram (Tg) is one million metric tons). These fluxes of Nr, and the Nr sources, sinks and transfers within the air, land and water compartments are presented in Table 1.

In the United States, human activity results in about 29 Tg N per year being added to the environment from all sources. In comparison, natural ecosystems add about 6.4 Tg N per year. Human activities control the introduction of Nr into the US (Figure 1).

Consequences, Impacts and Metrics for Nr

The best and most important consequence of Nr is food production in the US and global food security. There are, however, numerous negative consequences from anthropogenic Nr, including photochemical smog, atmospheric particulate loading, ecosystem fertilization, acidification, and/or eutrophication, greenhouse effect and stratospheric ozone depletion. But mitigating risk from these factors is difficult because one reactive N-containing molecule can contribute to all of these effects as a consequence of the nitrogen cascade (Figure 2). Nitrogen is a dynamic element easily transformed from one species to another and is transported rapidly through and between ecosystem reservoirs. These characteristics make it an especially challenging element to control.

Because nitrogen is both a critical resource and also a contributor to a number of environmental problems, it is imperative to understand how to reduce the risks to society while also providing the materials, food and energy required by society.

Various approaches can be used to prevent, eliminate, reduce, or otherwise manage risk. Understanding the environmental impacts of Nr can inform decisions on how best to manage nitrogen risks. There are two main approaches to this problem – traditional impacts and ecosystem services.

Traditional impacts include global warming, eutrophication, ecotoxicity, human health (cancer and non-cancer), acidification, smog formation, and ozone depletion, among others. Sometimes these impacts can be expressed in collective metrics. Collective metrics have the considerable advantage of defining a straightforward framework within which environmental standards can be derived that are protective of human health and the environment, the principal mission of the USEPA. Such metrics also encourage evaluation of damage from collective sources, as long as the characterization metric used is genuinely representative of the impact of a given contaminant. Thus, for example, the total impact of acidic gases such as SO_2 and NO_x on the acidification of

watersheds can be expressed as a common metric. However, metrics for human health are generally not as simple to characterize nor are there defined end points, thus the mechanism of toxicity, number of individuals affected, value of lost workdays, medical treatment costs, and value of human lives lost may all be used.

The ecosystem services approach complements traditional impact characterizations by assessing causative contaminant emissions. It considers how a specific service provided by one or more ecosystems or the corresponding causative functions (e.g. categories such as climate change, nutrient cycling, and food production) is impaired. The attractiveness of this approach lies in its recognition that the health of humans and the environment are inextricably linked. Less clear, in some cases, are ways in which to measure and monitor these impacts.

Both ways of expressing nitrogen impacts have value. Traditional categories (i.e., effects based) provide a readily adaptable framework for regulation. Function-based categories (i.e., services based) provide a richer context for the complex connections among Nr inputs and transformations. Further, their impacts on human well-being and dollar-based impacts can identify those effects that have the greatest damage costs to society. Using multiple metrics may provide a clearer picture of priorities for action, identify effective control points for reducing Nr impacts, and provide insights into more effective regulatory strategy.

Tradeoffs Among Nr Risk Reduction Options are Complex

Once the foreseeable impacts are understood and the suite of benefits associated with various risk reduction options described, then managers can consider trade-offs. Risk reduction integration provides an intellectual framework that allows managers to make informed decisions about which benefits may need to be relinquished for other benefits when not all the desired benefits can be achieved. For example, limiting nitrogen fertilizer application to reduce risks from Nr applied to agro-ecosystems risks reduced yields and higher commodity prices, which in turn may result in expansion of crop production area at the expense of natural wetlands, grasslands, and forests.

Measurement of Nitrogen in the Environment

What you measure determines both what you do and how you gauge success or failure. Most regulations set limits or specify control technologies for specific forms of nitrogen without regard to the ways in which nitrogen is transformed once introduced into the environment. Normally regulations also require some form of monitoring to document compliance. Monitoring of these specific forms of nitrogen is not enough. There is a need to measure, compute, and report the total amount of Nr, in appropriate units, present in impacted systems in appropriate units because one form of Nr can be quickly converted to other forms.

The impacts of reactive nitrogen often can be expressed as the dollar costs of damages, the cost of remediation or substitution, or the cost/ton of remediation for each form of reactive nitrogen. Damage costs do not always scale as tons of reactive nitrogen released into the environment. If damage costs rather than tons of nitrogen were utilized as a metric, the full implications of the cascade, and the setting of priorities for intervention might differ. Similarly if human mortality and morbidity are the metrics used, priorities for Nr releases could be very different.

Integrated Risk Reduction Strategies for Nr

Typically, quantitative risk assessment; technical feasibility; economic, social and legal factors; and additional benefits of the various control strategies contribute to the development of a suite of risk reduction strategies from which managers select an approach.

Control Strategies for Nr

There are several ways in which the release and control of Nr in the environment are approached. In general these can be classified as follows:

- Transformation—in which one form of nitrogen is converted to another form (e.g. nitrification, denitrification),
- Removal—in which Nr is sequestered from impacting a particular resource (e.g. ion exchange)
- Source limitation—in which the amount of Nr introduced into the environment is lowered (e.g. lower fertilizer application rates, controls on NO_x generation)
- Improved use efficiency—in which the efficiency of production that is dependent on Nr is improved (e.g. increased grain yields for lower Nr applied, or reduced NO_x from more efficient energy sources)
- Improved practices—in which the flux of Nr that creates an impact is lowered through better management practices (e.g. on-field agricultural practices, control of urban runoff, controlled combustion conditions)
- Product substitution—in which a product is developed or promoted which has a lower dependency on Nr (e.g. switchgrass instead of corn grain as a feedstock for ethanol)

Effective management of Nr requires combinations of these approaches; no one approach is a perfect alternative for controlling Nr in the environment.

Management of Nr in the Environment

Generally speaking, US environmental policy employs four mechanisms for the management of contaminants in the environment:

- Command-and-Control—in which permitted limitations on emissions, as promulgated under various statutes, are issued. Violations may result in the assessment of penalties.
- Government-based programs for effecting a policy, such as directed taxes, price supports for a given commodity, subsidies to bring about a particular end, and grants for capital expansion or improvement.
- Market-based instruments for pollution control in which cap and trade markets are used to bring about a desired policy end, often at reduced overall cost.
- Voluntary programs in which desired ends are achieved using private or government-initiated agreements or through outreach and education.

An integrated approach to the management of Nr must of necessity use a combination of mechanisms, each most appropriate to the nature of the problem at hand, that are supported by critical research on reducing the risks of Nr, and reflective of an integrated policy that recognizes the complexities and tradeoffs associated with the nitrogen cascade. Control at one point in the cascade may be more efficient and cost effective than control or intervention at another point. This is why understanding the nature and dynamics of the N cascade is so critically important.

Major Findings and Recommendations

The following are some of the Committee's major draft recommendations.

1. There is a pressing need to encourage an adaptive, precision-conservation approach to terrestrial nutrient management, crop production, animal management, and agricultural and urban runoff. It is possible to reduce excess flows of Nr into streams, rivers, and coastal systems by approximately 20% **(~1** Tg N per year.**)** through improved landscape management without undue disruption to agricultural production and human lifestyles and economies. This would include activities such as using wetland management (e.g., USDA Wetlands Protection Program), improved tile-drainage systems and riparian buffers on crop land, and implementing storm water and non-point source management practices (e.g., EPA permitting and funding programs).

   It is also possible to increase crop N-uptake efficiencies by up to 25% over current levels through a combination of knowledge-based practices and advances in fertilizer technology (such as controlled release). The net reduction would be somewhat less as some duplication of efforts is represented in reducing excess Nr flows and increasing N-uptake efficiencies. However, the critical conclusion is that crop output can be increased while reducing total Nr by up to 20% of applied artificial Nr, amounting to ~**2.4** Tg N per year below current levels of Nr additions to the environment. These are appropriate targets with today's available technologies; further progress is possible.

2. The Clean Air Act (1970) and its Amendment (1990), have resulted in NOx emissions that are <50% of what they would have been without the controls. While this is an admirable accomplishment, there is still a way to go, as NOx emissions are still an order of magnitude greater than at the beginning of the 20th century and, as a consequence, there are still negative impacts on both people and ecosystems.

   We recommend that the EPA expand its NOx control efforts from the current reductions of emissions of passenger cars and power plants to include other important unregulated mobile and stationary sources. Notable NOx emitters include heavy-duty on-road and all off-road mobile sources (including rail and marine), as well as currently uncontrolled electricity generation and industrial processes. Well-regulated electricity generating units and light duty vehicles currently eliminate ~90% of the NOx they would otherwise emit. Instituting 90% reductions for the major, currently uncontrolled sources would reduce annual emissions by about **2** Tg N per year. This may be sufficient to bring most of the US into compliance with the current O_3 NAAQS, but may still leave several ecosystems with more Nr than the critical load.

It is vitally important that the implementation of these controls not result in additional emissions of N_2O and NH_3 to the atmosphere, which would just change one N-related problem to another.

3. In spite of gains made over the last several decades in lowering the amount of NOx emitted from stationary and mobile combustion sources, the total amount of Nr released into the atmosphere has remained relatively constant. This is related largely to the essentially unregulated release of ammonia from livestock operations (mostly due to increasing poultry and swine production), which have expanded significantly. Ammonia emissions from livestock production have increased ~30% since 1970. We suggest a goal of decreasing livestock-derived ammonia emissions to approximately 80% of 1990 emissions, a decrease of **0.5** Tg N per year (by a combination of Best Management Practices and engineered solutions). This will reduce PM2.5 by ~0.3 $\mu g/m^3$ (2.5%) and improve health of ecosystems by achieving progress towards critical load recommendations. Additionally we recommend decreasing ammonia emissions derived from fertilizer applications by 20% (decrease by ~**0.2** Tg N per year.).

4. National loadings of Nr to the environment from public and private wastewater point sources are relatively modest in comparison with other releases to the environment, but can be important local sources with associated impacts. In most cases Nr ultimately finds its way into municipal and private sewers and treatment systems where, irrespective of its initial chemical form, it is partially or completely nitrified. Subsequent engineered complete denitrification processes (including tertiary wastewater treatment, engineered or restored wetlands, and algae production for biofuels) can convert the nitrate to only N_2. Federal and State assistance programs directed at construction of treatment plants are an important Nr control policy in the US. The committee recommends that a high priority be assigned to nutrient management through a targeted construction grants program under the CWA. The committee believes that **0.5 to 0.8** Tg N per year can be saved from Nr inputs to the environment.

5. Acreage devoted to corn production has increased about 10% for corn based ethanol production, with nearly one-third of the crop being devoted to bioethanol production. Current policy calls for bioethanol to expand to 15 billion gallons for corn-based ethanol and 36 billion gallons of bioethanol from all sources by 2022. We expect fertilizer nitrogen to increase by at least 10% **(0.5** Tg N per year**),** initially to meet biofuel feedstock crop demand. Strategies to increase N-uptake efficiencies and strategies to reduce N losses must be implemented across corn and other N intensive biofuel crops.

   N_2O in the atmosphere is also increasing. For additional production of liquid biofuels beyond the grandfathered amount in the 2007 Energy Independence and Security Act (EISA), EPA has the power to exercise some controls on N_2O emissions through the life cycle greenhouse gas accounting requirements.

   In the absence of Nr controls and a failure to implement best practices, current biofuels policies will make it extremely difficult to reduce Nr releases to soils, water and air. Integrated management strategies will be required. In this regard, we endorse Section 204 of EISA which requires that after 3 years and then every 3 years thereafter, the EPA

Administrator, the Secretary of Agriculture, and the Secretary of Energy shall report to Congress on the impact of the Clean Air Act requirements related to environmental issues, resource conservation issues, and the growth and use of cultivated invasive and noxious plants. (http://www.ethanol.org/pdf/contentmgmt/Full_Text_of_HR6.pdf)

6. The current air pollution indicator for oxides of nitrogen, NOx, is an inadequate measure of reactive nitrogen in the atmospheric environment. We recommend that the inorganic reduced nitrogen (ammonia plus ammonium) and total oxidized nitrogen (NOy) be monitored as indicators of total chemically reactive nitrogen. The basis for the recommendation is that inorganic reduced nitrogen has environmental impacts equivalent to the current criteria air pollutants.

7. There is an urgent need to improve and maintain foundational data required to track sources of Nr and Nr loads in the environment. Specific data needs include: the rationalized and geospatially defined fertilizer use data; improved estimates of nitrogen fertilizer efficiency and its variation based on estimates from production-scale fields for the major crops and cropping systems; and improved monitoring and estimates of wet/dry Nr deposition and its transformation and transport on land and in water.

8. What is managed depends on what is measured, and because Nr undergoes multiple chemical transformations as it cascades through multiple media and ecosystems, impacts and intervention points are difficult to determine. There are many metrics for evaluating and prioritizing Nr impacts. The most widely used traditionally measure has been mass of nitrogen by chemical species, but one can also measure damage costs of impacts, or replacement and mitigation costs or human health measures. The use of multiple metrics may provide a fuller picture of the impacts of reactive nitrogen and improve the setting of priorities.

The actions recommended above would decrease the amount of Nr entering the environment by ~7 Tg N/yr, or about 25% of the anthropogenic Nr created each year in the US. Other actions could be taken, and all actions need to take into consideration an over-arching finding of the committee—as the amount of reactive nitrogen released to the environment grows, more effective integration of strategies that work across media, address multiple problems and avoids unintended adverse consequences is necessary to reduce costs and create more enduring solutions.

The Committee's recommended actions have real economic costs. Trade-offs will be made both within and between recommendations. For example, treating nitrate with engineered wetlands, that provide additional benefits, such as the production of algae for biofuels, may prove to be more cost effective than traditional tertiary treatment. Similarly, where reducing ammonia emissions from animal feeding operations can be paired with the recovery of methane for fuel then overall costs should be lower and greenhouse gas emissions will also be reduced.

The Committee's recommendations represent realistic intermediate targets based on current demands and technologies. There are and will be opportunities to go beyond these recommendations. Developing these opportunities will be critical given the growing demand from population and economic growth for food- and fiber-production and energy use.

Concluding Statement

Fossil fuel combustion and food production have significantly increased the introduction of Nr into the US environment and, while there are tremendous benefits, there are also tremendous damages to the health of both ecosystems and people. Optimizing the benefits of Nr while minimizing its problems will require an integrated nitrogen management strategy that not only involves EPA, but also other federal agencies (e.g., USDA, DOE, NOAA), state agency managers, the private sector and a strong public outreach program.

Table 1. Reactive nitrogen fluxes for the USA, Tg N in 2002*

Nr inputs to Atmospheric compartment		
N_2O-N emissions		0.8
agriculture - Soil management	0.5	
*fossil fuel combustion - transportation N_2O	0.1	
Miscellaneous	0.1	
NHx-N emissions		3.1
agriculture: livestock NH_3-N	1.6	
agriculture: fertilizer NH_3-N	0.9	
miscellaneous	0.6	
NOx-N emissions		6.2
*fossil fuel combustion - transportation NOx	3.5	
*fossil fuel combustion - utility & industry NOx	1.9	
miscellaneous	0.9	
Nr inputs to Terrestrial compartment		
atmospheric N deposition	6.9	
*N fixation in cultivated croplands	7.7	
*N fixation in non-cultivated vegetation	6.4	
*N import in commodities	0.2	
*N fertilizer use on farms & non-farms	10.9	
*non-fertilizer uses	4.2	
manure N production	6.0	
human waste N	1.3	
Nr inputs to Aquatic compartment		
surface water N flux		4.8

*these fluxes represent injection of new Nr into the USA

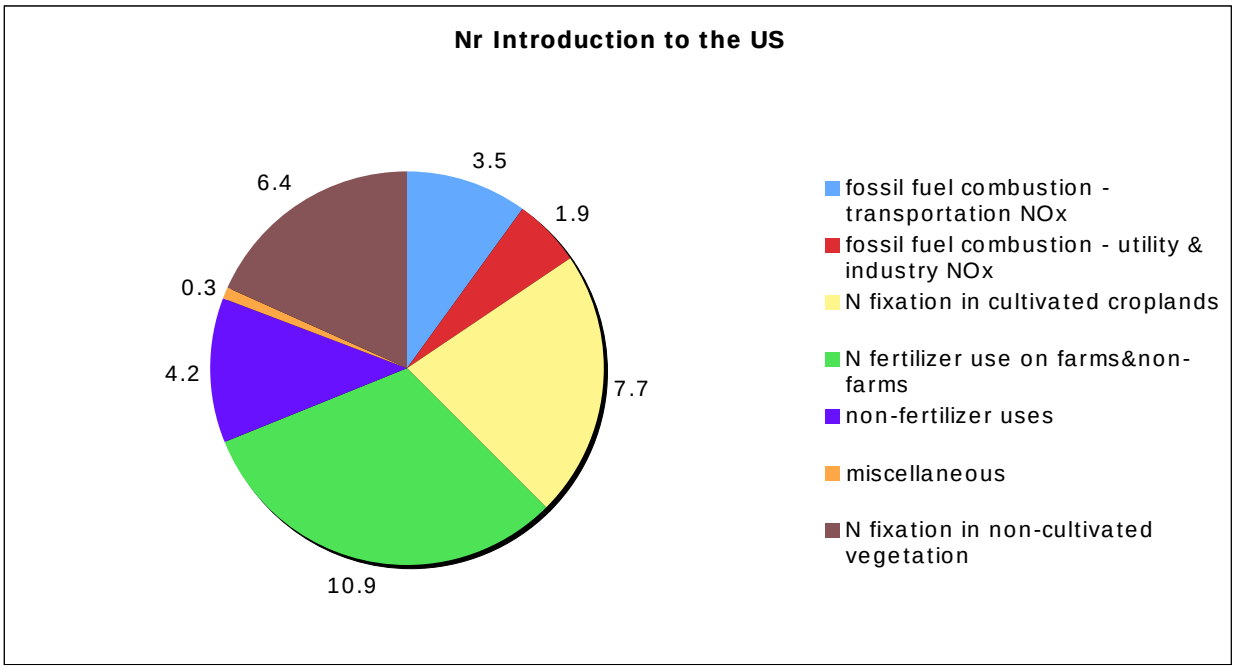

Figure 1: New Nr introduced into the US, 2002, Tg N.

Note that the numbers from the table do not all match up with the figure because some recycled Nr is included in the table (livestock, manure, and human sewage).

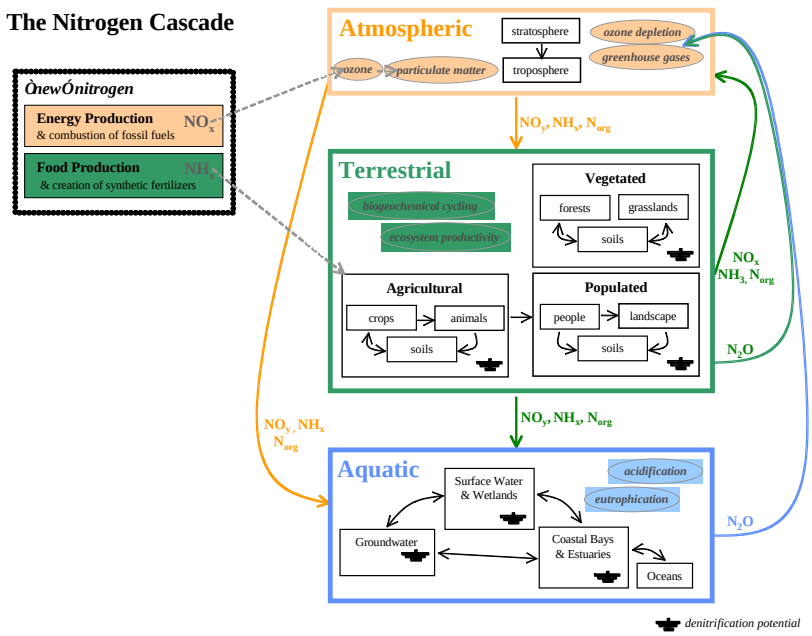

Figure 2: The Nitrogen Cascade: The popular concept of the nitrogen cascade highlights that once a new Nr molecule is created, it can, in sequence, travel throughout the environment contributing to major environmental problems (Galloway et al., 2003). This adaptation of the cascade was developed by the Integrated Nitrogen Committee to provide a context for considering nitrogen-related issues and ecosystem effects in the US. To consider the cascading effects of Nr in the US, we examine the relative sizes of the various Atmospheric, Terrestrial, and Aquatic compartments where Nr is stored, and the magnitudes of the various flows of nitrogen to-, from-, and within them. The nitrogen cascade concept implies the cycling of Nr among these compartments. The important process of denitrification is the only mechanism by which Nr is converted to chemically inert N_2, 'closing' the continuous cycle.

The "new" nitrogen box depicts the two primary sources by which Nr originates, energy production and food production, and where they enter ecosystems. Energy production includes both fossil fuel and biofuel combustion. Food production includes N fertilizer produced in the US, cultivation-induced biological nitrogen fixation in the US, production of animals and crops in the US for human consumption, and imports of N-containing fertilizer, grain and meat to the US.

The Atmospheric compartment indicates that tropospheric concentrations of ozone, particulate matter and nitric acid are increased due to NOx emissions to the atmosphere. The ovals illustrate that the increase in N_2O concentrations, in turn, contribute to the greenhouse effect in the troposphere and to ozone depletion in the stratosphere. Except for N_2O, there is limited Nr storage in the atmosphere. Losses of Nr from the Atmospheric compartment include NOy (which includes HNO_3 and particulate nitrate), NHx, and Norg deposition to Terrestrial and Aquatic

ecosystems of the earth's surface. These depositions contribute to both acidification and eutrophication of land and water. There is little potential for conversion of Nr to N_2 via denitrification in air.

The Terrestrial compartment depicts Nr entering agricultural lands via food production and is introduced to the entire terrestrial landscape via atmospheric deposition. Within 'agricultural' regions there is cycling among soils, crops and animals, and then a transfer of Nr as food to 'populated' regions, from which there are Nr losses (e.g, sewage, urban runoff). The ovals showing 'ecosystem productivity' and 'biogeochemical cycling' reflect that Nr is actively transported and transformed within the Terrestrial compartment, and that as a consequence there are significant impacts on ecosystem productivity due to fertilization and acidification, often with resulting losses of biodiversity. There is ample opportunity for Nr storage in both biomass and soils. Losses of Nr from the Terrestrial compartment occur by leaching of NOy, NHx and Norg to Aquatic ecosystems and by emissions to Atmospheric ecosystems as NOx, NH_3, Norg, and N_2O. There is some potential for conversion of Nr to N_2 via denitrification in the landscape.

The Aquatic compartment shows that Nr is introduced via leaching from Terrestrial ecosystems and via deposition from Atmospheric ecosystems. Connected with the hydrological cycle, there are Nr fluxes downstream with ultimate transport to coastal systems. Within the Aquatic compartment, the ovals highlight two significant impacts of waterborne Nr—acidification of freshwaters and eutrophication of coastal waters. Except for Nr accumulation in groundwater reservoirs, there is limited Nr storage within the hydrosphere. Losses of Nr from the Aquatic compartment are primarily N_2O emissions to Atmospheric ecosystems. There is a very large potential for conversion of Nr to N_2 via denitrification in water and wetlands.