Comments on First External Review Draft of "Integrated Science Assessment for Oxides of Nitrogen and Sulfur: Environmental Criteria"

Prepared for the
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Executive Summary

The effects of acidification and nitrogen-nutrient deposition on ecosystems are the primary focus of the draft Integrated Science Assessment (ISA). Although the U. S. Environmental Protection Agency (EPA) is combining the review of oxides of nitrogen (NOx) and sulfur (SOx) welfare effects in this one document, these Air Improvement Resource, Inc. (AIR) comments concentrate on the science that will influence the NOx secondary standard decision. There was substantial evidence of NOx-related acidification of freshwater streams and lakes and eutrophication of estuaries and coastal waters in the last review completed in 1996. However, at that time, the Administrator concluded that adoption of a nationally-uniform secondary standard would not be an effective way of approaching these problems.

In the last decade there has been substantial new information on acidification effects on terrestrial and aquatic ecosystems and eutrophication effects of excess nitrogen. There have also been major reductions in man-made NOx and SOx emissions as well as control programs put in place for additional NOx and SOx reductions in the coming decades.

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

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

- There are both technical and legal considerations in choosing an appropriate indicator; the ISA needs to include a short discussion of the legal questions since the Clean Air Act presupposes nationally uniform primary and secondary national ambient air quality standards (NAAQS).

- The ISA should evaluate both reduced and oxidized forms of reactive nitrogen equally. The discussion needs to start with a description and understanding of the natural N cycle.

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

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

- The discussion of NOx emissions from soil and lightning is missing important papers that demonstrate these sources result in significantly higher emissions than noted in the ISA

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

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

- If deposition-based secondary standards are to be considered, a discussion of the quality and completeness of deposition measurements should be included in the ISA.

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

- Issues with the critical loads approach need to be explicitly discussed in the body of the ISA. For example, although the Europeans have been developing critical loads as a tool for evaluating adversity and setting policy, they do it in a cost-benefit framework. This is not consistent with the U. S. NAAQS paradigm.

- The implications of the fact that many sources contribute to eutrophication need to be discussed. .

    - Eutrophication is not primarily an air pollution problem; it involves not only combustion sources of NOx, but also agricultural practices, fertilizer use, treatment of human waste, and treatment of animal waste.

    - The wide range of atmospheric contributions, sensitivities of different water resources, and contributions from non-atmospheric sources makes using a national ambient air quality standard to address eutrophication problematic.

Based on the comments below, the draft ISA needs extensive revision before it can serve as the basis for policy decisions regarding secondary NAAQS for NOx and SOx.

Introduction

The draft "Integrated Science Assessment for Oxides of Nitrogen and Sulfur: Environmental Criteria"[1] (ISA) is designed to be a concise synthesis and evaluation of the most policy-relevant science for use in EPA's decision-making process concerning appropriate secondary air quality standards for oxides of nitrogen (NOx) and sulfur (SOx). Since NOx and SOx emissions contribute to ambient particulate matter (PM), EPA will be considering some NOx and SOx-related welfare effects such as visibility reduction in the review of the secondary PM standards. The effects of acidification and nitrogen-nutrient deposition on ecosystems are the primary focus of this draft ISA. Although EPA is combining the review of NOx and SOx effects in this one document, these AIR, Inc. comments concentrate on the science that will influence the NOx secondary standard decision.

In October 1996, in the previous review of the NOx secondary standard, EPA decided to set the secondary standard identical to the primary standard. Although there was substantial evidence of NOx-related acidification of freshwater streams and lakes and eutrophication of estuaries and coastal waters, the Administrator concluded that adoption of a nationally-uniform secondary standard would not be an effective way of approaching these problems.[2] The Administrator indicated that the decision would be re-visited in the next review and that, meanwhile, the Title IV program that addresses acid deposition and other national and regional programs will be reducing NOx emissions. At that time, consideration was given to an acid deposition standard, but CASAC[3] and EPA staff[4] agreed that the available scientific information did not provide an adequate basis for standard setting purposes.

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

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[1] U, S. Environmental Protection Agency, First External Review Draft of "Integrated Science Assessment for Oxides of Nitrogen and Sulfur: Environmental Criteria," EPA/600/R-07/145A, December 2007; U, S. Environmental Protection Agency, First External Review Draft of "Annexes for the Integrated Science Assessment for Oxides of Nitrogen and Sulfur: Environmental Criteria," EPA/600/R-07/145bA, January 2008.
[2] 61 Federal Register 52852, October 8, 1996.
[3] G. T. Wolff, CASAC closure letter for the 1995 OAQPS Staff Paper, addressed to U. S. EPA Administrator Carol Browner, August 22, 1995.
[4] U. S. Environmental protection Agency, Review of the National Ambient Air Quality Standards for Nitrogen Dioxide: Assessment of Scientific and Technical Information, EPA-452/R-95-005, September 1995.
[5] U. S. Environmental Protection Agency, Acid Deposition Feasibility Study Report to Congress, 1995
[6] National Research Council, Air Quality Management in the United States, National Academy Press, Washington DC, 2004.

In the decade and more since the last NOx Criteria Document (1993),[7] there has been substantial new information on acidification effects on terrestrial and aquatic systems and eutrophication effects of excess nitrogen. There have also been major reductions in man-made NOx and SOx emissions as well as control programs put in place for additional NOx and SOx reductions in the coming decades.

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

General Comments

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

While the choice of indicator is ultimately a policy decision, the ISA needs to adequately cover the science that would support that policy choice. Since a separate secondary standard with different indicator, form, averaging time, concentration, and/or allowed frequency of occurrence from the primary standard has not been in place for either SOx or NOx, the form and usefulness of alternative prospective indicators will be the important threshold question in the review. In fact, the December 2007 Review Plan indicates that the issue of appropriate indicators is central to the review of the standards.[8] The various options for indicator are not obvious from reading the draft ISA and so it is impossible to tell if it adequately covers the science behind each option. Therefore, an explicit listing and discussion of potential indicators needs to be included in the ISA. In CASAC comments on the draft review plan, members of the panel indicated that analysis of the many options for regulating deposition needs to be part of the ISA and requested that the technical foundations for use of potential indicators should be presented up front in the ISA.[9]

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

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

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

[9] See CASAC letter to Administrator Johnson EPA-CASAC--08-003, November 29, 2007, at pages 31, 36, and 41.

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

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

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

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

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

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

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

[10] NRC 2004 at page 312.

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

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

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

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

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[11] C. Luo, C. Zender, H. Bian, and S. Metzger, "Role of ammonia chemistry and coarse mode aerosols in global climatological inorganic aerosol distributions," Atmos. Environ., **41**, 2510-2543 (2007).
[12] ISA at page 4-2.
[13] N. Gruber and J. Galloway, An Earth-system perspective of the global nitrogen cycle, Nature , **451**, 293-296 (2008).
[14] CASAC 08-003 at page 28.
[15] See P. Hogberg, "Nitrogen impacts on forest carbon," Nature, **447**, 781-782 (2007); F. Magnani, et al, "The human footprint in the carbon cycle of temperate and boreal forests," Nature , **447**, 848-450 (2007); K. Nadelhoffer, et al., "Nitrogen deposition makes a minor contribution to carbon sequestration in temperate forests," Nature, **398**, 145-148 (1999).

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

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

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

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

For example, the kind of sources, spatial distribution of sources, trends in emissions, and potential effects of deposition are different between reduced and oxidized forms. The current draft focuses primarily on the oxidized forms. However, NOx emissions from fossil fuel combustion have been decreasing for several decades and will continue to decrease for the next several decades due to controls that are already "on the books." In contrast, the sources of reduced nitrogen (such as fertilizer use) are still increasing as noted below in the specific comments on ammonia emissions inventories. Therefore, additional detail on the sources and trends in reduced N emissions should be added and indicators that include reduced nitrogen should be explicitly evaluated. Several members of the CASAC panel, in comments on the draft review plan, agreed that reduced nitrogen needs to be fully included in the ISA.[19]

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

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

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

[18] ISA at page 4-43.

[19] See CASAC –08-003 at pages 4, 7. 17, 23, 36, and 41.

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

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

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

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

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

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

Specific Comments

Chapter 1 – Introduction

Both the Review Plan (at page 3-2) and the ISA (at page 1-2) include lists of framing questions that the ISA and, ultimately, the current review must answer. The ISA, however, does not address these questions explicitly. Questions such as "Does the available information provide support for considering joint standards or are separate standards appropriate?" and "What forms of alternative standards are supported by the information and what are the uncertainties and limitations in that information?" are some of the questions that need to be explicitly addressed in the ISA. In section 1.1.2, the scientific basis for the administrator's decision in the previous review should be summarized. As noted above, although there was substantial evidence of NOx-related acidification of freshwater streams and lakes and eutrophication of estuaries and coastal waters, the Administrator concluded that adoption of a nationally-uniform secondary standard would not be an effective way of approaching these problems.[20] The Administrator indicated that the decision would be re-visited in the next review and that, meanwhile, the Title IV program that addresses acid deposition and other national and regional programs will be reducing NOx emissions.

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

Clarity in the nomenclature is imperative to avoid confusion

The introduction to Chapter 2 indicates that the ISA will use the term NOx to refer to all forms of oxidized nitrogen compounds in contrast to the nomenclature typically used in the scientific community wherein NOx refers to the total of just two of the oxides of nitrogen, NO and NO_2. However, at numerous places in the ISA there is confusion since NOx is used as it used in the general literature. For example, the description of nitrogen chemistry at pages 2-3 to 2-7 uses the nomenclature where NOx + NOz = NOy, where NOy is the total of all oxidized nitrogen. To avoid such confusion, it is important to be very specific throughout the document to make sure that the chemical forms of nitrogen included in a given term or measured quantity are clear.

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

Chapter 2 notes that the ISA properly includes discussion of ammonia as a precursor of NOx in gas phase reactions as a source of ammonium that neutralizes acids as well as a separate actor in nitrification of terrestrial and aquatic ecosystems. Because of the importance of emissions, transport, transformations, and deposition of reduced forms of nitrogen, the ISA should include a review and discussion of reduced N throughout at the same level of detail as oxidized nitrogen.

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[20] 61 Federal Register 52852, October 8, 1996

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

While the draft chapter includes several pages of description of prominent gaseous and heterogeneous reactions of NOx species, the reader does not get a good feel for the relevant time and spatial scales involved between emission and deposition. Since a knowledge of the spatial scale of influence (or footprint, as it might be called) of relevant emissions will be key input into the consideration of potential secondary standards, it is important to include additional discussion of the spatial scale. There are differences between the N deposition footprint due to NOx emissions emitted at ground level in urban areas generally in combination with VOC, and NOx emitted from tall stacks in combination with SO_2, and reduced nitrogen emitted at ground level in rural areas.[21] For example, a recent study by Elliot, et al. 2007[22] of nitrogen isotopes in wet nitrate deposition reported that wet nitrate at 33 sites in the Midwestern and Northeastern U. S. is strongly associated with NOx emissions from stationary sources.

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

As indicated in Figures 2.2-1 and 2.3-1 and in text of Sections 2.2 and 2.3, the atmospheric chemistry of SOx and NOx is quite complex. For the purposes of the ISA, the draft should list and discuss the major sources of non-linearity in these coupled atmospheric oxidation processes. While NOx and SOx emissions that are emitted to the atmosphere will eventually be removed from the atmosphere, factors which will influence these processes in a non-linear way will shift where and when the materials are deposited, whether they are removed from the system or are re-cycled, and whether they are partitioned into different forms that may be more or less harmful or beneficial. Such differences are important and will influence the extent that current control programs reduce harmful deposition in sensitive areas and thereby affect potential adverse effects.

Chapter 2 includes discussion of heterogeneous chemistry involving halogen-radical reactions that may influence NOx and SOx oxidation especially in marine boundary layers. In the SOx section, it is acknowledged that the absolute magnitudes and relative importance of these processes are poorly understood. If this is true for the similar NOx reactions, it should be so stated. The extent to which the heterogeneous chemistry discussed in Chapter 2 is included in the atmospheric models discussed in Chapter 3 should be explicitly discussed.

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

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[21] See also comment by CASAC panelist at page 44 of CASAC-08-003.

[22] E. Elliot, C. Kendall, S. Wankel, D. Burns, E. Boyer, K. Harlin, D. Bain, and T. Butler, "Nitrogen isotopes as indicators of NOx source contributions to atmospheric nitrate deposition across the Midwestern and Northeastern United States," Environ. Sci. Technol., **41**, 7661-7667 (2007).

Chapter 2 also covers the atmospheric measurements of NOx and SOx species. The ISA acknowledges that routine NO_2 measurements in the monitoring network are subject to unknown and varying interferents from other nitrogen oxide species, so they are not a measure of either NO_2 or total oxidized nitrogen. Because of these concerns, there is a variable degree of positive bias in the data. The Health Criteria NOx ISA notes that this bias may be roughly 20 to 25 %.[23] Such a large positive bias is not acceptable. First, the specific techniques used to convert NO_2 to NO currently in use throughout the country should be documented by EPA and the States and the biases and interferences in those techniques should be carefully evaluated. Second, it is incumbent on the Agency to develop more specific NO_2 measurement techniques. The ISA also discusses research-type instruments or instruments under development that can measure NOy or individual NOx species noting issues and problems suggesting they are not ready for routine monitoring. Yet the ISA concludes that:

> "For the purposes of identifying and quantifying the atmospheric concentrations and deposition totals causing ecological effects, these measurement techniques and sampling frequencies are fully adequate."

This statement is too general and is not supported by the material in the ISA. In addition, a decision regarding adequacy is a policy decision; the ISA should stick to the facts concerning the monitoring methods. If concentration-based indicators are considered for secondary standards, there needs to be an explicit discussion of the techniques available for those indicators be they specific gaseous or particulate species or combinations and the possible utility of those indicators. For example, the ISA discusses measurement techniques for NOy, total oxidized nitrogen. However, the atmospheric concentration of total oxidized nitrogen would not be meaningful for evaluating ecological risk, since it a measure of a mixture of compounds which each have their own inherent deposition characteristics and possibly different effects in ecosystems. In addition, the atmospheric concentration of any species or combination of species would be an inappropriate indicator because it does not include wet deposition

The quality and completeness of deposition measurements should be discussed

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

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

[24] U. S. Environmental Protection Agency, Clean Air Status and Trends Network (CASTNET) 2006 Annual Report, November 2007.

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

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

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

For example, the chapter discusses both trends in emissions and ambient measurements for SOx in Section 3.2 but not for NOx in Section 3.1. The reader should not have to go to a table in the annexes to find the breakdown of NOx or ammonia emission sources. The trends in reduced and oxidized N are also important to include in the body of the ISA.

To enable direct comparisons of various emission sources and fluxes in the N cycle, the quantities should be presented and compared on a consistent basis. Because of the potential use of N deposition as an indicator metric, the emission data should be reported on a mass of nitrogen basis. For example, NOx ($NO + NO_2$) emissions are typically reported as mass of NO_2. This results in a very misleading comparison between the mass of NOx and NH_3 emissions in Table AX2-1 when not presented on a mass of N basis.

In Section 3.3 the text indicates that the anthropogenic emissions of ammonia show a strikingly different pattern from that of NOx or SOx. However, that pattern is not shown. It should be. If emission trend data for reduced nitrogen is not available, trends in fertilizer use in the U. S. should be presented and discussed as a reasonable surrogate. For example, historic trends in fertilizer consumption in North America[25] and projections of nitrogen fertilizer demand in North America until 2011/2012[26] are available. In contrast to the continuing reductions in NOx emissions, N fertilizer demand in North America is forecast to increase at 0.3 % per year through 2012.

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

For both SOx and NOx, regulatory programs already in place are projected to reduce emissions well below current levels. The ISA should document these reductions since they will occur with or without separate secondary standards.

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

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

The discussion of NOx emissions from soil and lightning is missing important papers demonstrating that these sources result in significantly higher emissions than noted in the ISA

There are NOx emissions from soils due to microbial processes, and fertilized agriculture produces the highest soil NOx emissions. The (Health) ISA references the Jaegle et al. 2005[27] study that used satellite observations of NO_2 columns to derive estimates of NOx emissions from fuel combustion, biomass burning, and soils. Jaegle et al. conclude that there is a significant role for soil NOx emissions at northern mid-latitudes during the summer, where they account for nearly half that of the fuel combustion source. The soil NOx estimated by Jaegle et al. was twice that in the inventory used in the GEOS-CHEM model. The Jaegle et al. estimate should be included in the ISA.

Another paper referenced in the ISA, Singh et al. 2007, found that lightning was a much larger contributor to NOx species in the upper troposphere over North America than previously thought. Singh et al. also refer to a new paper[28] in which the GEOS-CHEM model improved its prediction of the observational data in the upper troposphere by increasing the lightning source (NOx yield per flash) in the model by a factor of four. The Hudman et al. 2007 paper also provided boundary layer NOx measurements verifying a 50 % reduction in power plant and other NOx emissions over the eastern U. S. from 1999 to 2004. All these papers should be discussed in the ISA.

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

Section 3.5 discusses the uptake of N species to vegetation as an ecological sink. This section should include a reference to the fact that such interactions are not phytotoxic as noted in the conclusions on page 5-35. The complexities of these interactions is demonstrated by the fact that at low NO_2 concentrations (below 1 ppb) plants release NO_2 as documented in the material in Section 3.9.

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

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

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[27] L. Jaegle, L. Steinberger, R. Martin, and K. Chance, "Global partitioning of NOx sources using satellite observations: Relative roles of fossil fuel combustion, biomass burning, and soil emissions," Faraday Discussions, **130**, 407-423 (2005).

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

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

Section 3.7 which discusses ambient concentrations should include the trend in ambient NO_2 as well as the summary of current levels that is included. For example, Figure 1 below shows the distribution of annual average concentrations at all U. S. monitoring locations from 1970 through 2007. While the number of monitoring sites differs somewhat from year to year, a downward trend that has reduced ambient NO_2 concentrations by a factor of about three over the past 30-some years is evident.

Since most of the NO_2 monitoring is conducted in urban or suburban locations, the ambient trend is primarily indicative of emission reductions in and around urban areas. Since highway vehicle emissions are a major source of NOx in urban areas, a comparison of the ambient trend with the trend in highway vehicle emissions is illustrative. Figure 2 shows the trend in highway vehicle emissions on a gram per mile basis from 1970 through 2020 calculated with the U. S. EPA's MOBILE6 emission model, using the model's default inputs. The trend in NOx emissions from highway vehicles from 1970 to 2007 is very similar to the trend in ambient NO_2. Moreover, the downward trend in NOx emissions will continue well into the 2020s.

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

[29] ISA at page 3-20.
[30] ISA at page 3-50.

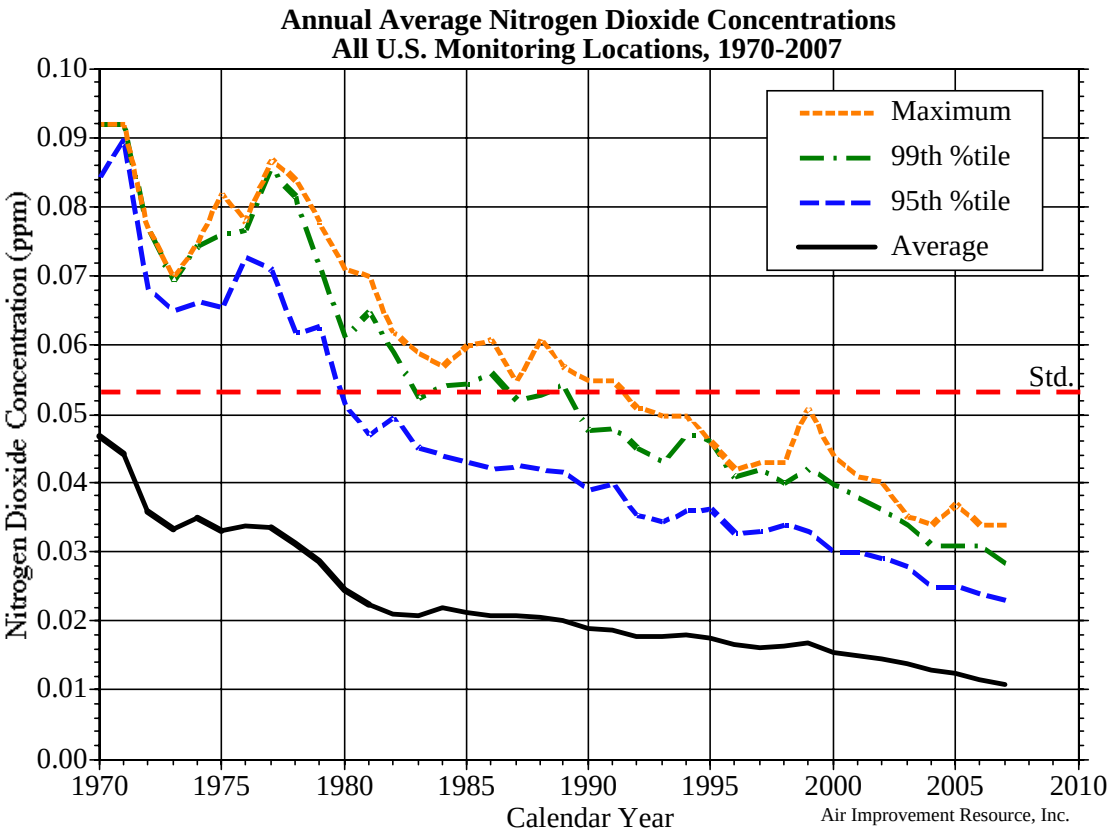

Figure 1

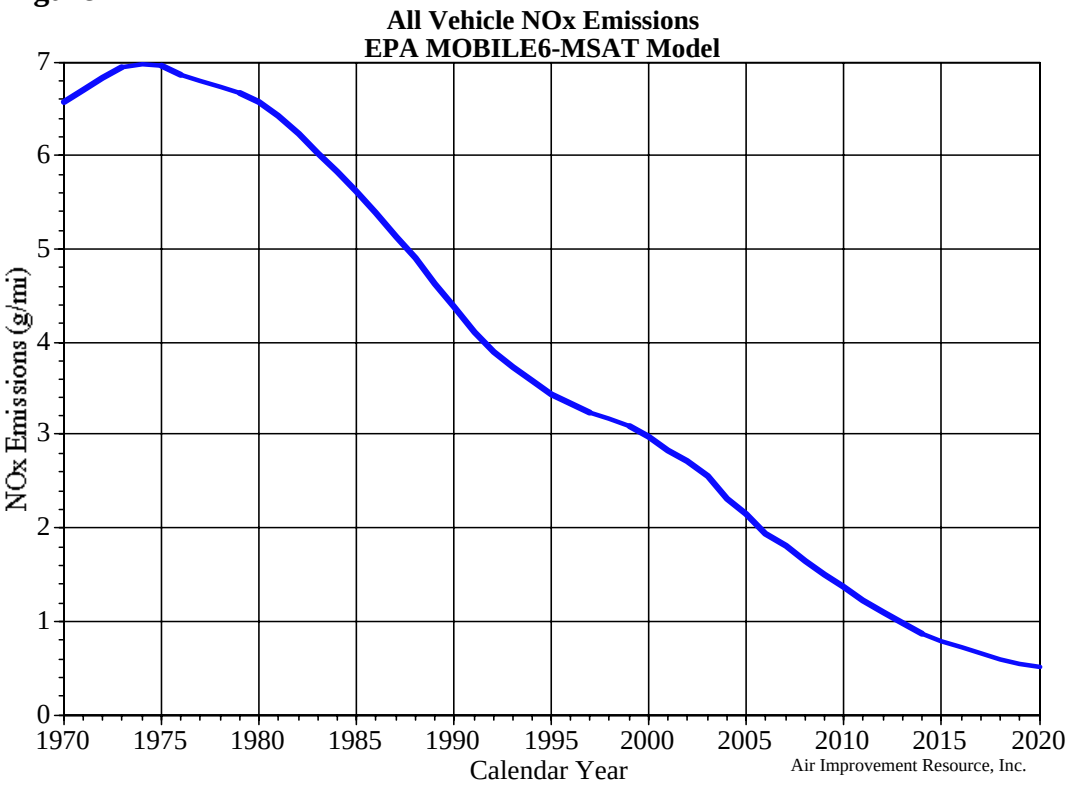

Figure 2

The other large source of NOx emissions is power plants. These sources have also been subject to substantial control and are scheduled for additional control under the Clean Air Interstate Rule. The ISA should document the reductions that have taken place to date and the reductions that are anticipated with current regulations for both motor vehicle and power plant emissions in the ISA to provide context for the interpretation of historic environmental data

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

It is not clear what Section 3.9 adds to the discussion

Section 3.9 is a detailed discussion of the fluxes of NOx species in Harvard Forest. It is not clear why this much detail is needed in the body of the ISA. The bottom line from this work is not clearly stated in the section. It does demonstrate the complexities of N interactions in the forest canopy and ecosystem. It also demonstrates that vegetation can be both a source of NO_2 at low ambient NO_2 concentrations and a sink for N species. The text indicates that direct NO_2 by plants is important to the plant but not a significant part of the overall N deposition to rural sites

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

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

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

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

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

The text refers to Table 3.11-2 which is just a list of sources in various categories to support the conclusion that NOx chiefly from fossil fuel combustion often dominates N pollution in the United States and comprises about 50 to 75 % of total N atmospheric deposition. The current draft ISA does not contain enough of the supporting data to determine whether this conclusion is warranted or not, given the contributions from fertilizer use and natural sources. The basis for the conclusion should be explicitly provided in the ISA. With the inclusion of the data suggested in these comments, the reader can make his or her own evaluation of the contribution of oxidized versus reduced nitrogen and the contribution of fossil fuel combustion to overall oxidized nitrogen deposition.

Although the policy relevant background concentration of NO_2 is far below current urban levels, an accurate definition and analysis of policy relevant background is necessary in the ISA.

The ISA defines a policy relevant background (PRB)[34] in Section 3.12 as those concentrations that would result in the United States in the absence of anthropogenic emissions in North America; i.e. leaving no room for NO_2 caused from human activities. This definition of PRB is flawed for several reasons. It omits the contribution from biogenic emissions due to agricultural activities and it omits consideration of the contribution from anthropogenic emissions in Mexico and Canada. While there can be

[31] CASAC 08-003 at page 27.
[32] B. Eder and S. Yu, " A performance evaluation of the 2004 release of Models-3 CMAQ," Atmos. Environ., **40**, 4811-4824 (2006).
[33] J. Davis and J. Swall, "An examination of the CMAQ simulations of the wet deposition of ammonium from a Bayesian perspective, Atmos. Environ., **40**, 4562-4573 (2006).
[34] ISA at page 3-51.

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

The ISA also errs by relying on one model rather than considering a range of results from both models and measurements. Although background is not as large an issue in the NOx secondary standard review as it is in the ozone review, studies included in the ISA show that EPA systematically underestimated natural NOx emissions and, hence, both background NO_2 in this review and background ozone in the ozone review. For example, the ISA indicates that the model used to estimate PRB for NO_2 (Mozart-2) has lower estimates (by about a factor of two) of soil NOx than the GEOS-CHEM model that was used to estimate PRB for ozone.[35] As noted above, the (Health) ISA references the Jaegle et al. 2005[36] study that used satellite observations of NO_2 columns to derive estimates of NOx emissions from fuel combustion, biomass burning, and soils. Jaegle et al. conclude that there is a significant role for soil NOx emissions at northern mid-latitudes during summer, with estimated soil NOx emissions twice that in the inventory used in the GEOS-CHEM model. Thus, the Mozart-2 model used in the ISA likely underestimates soil NOx at northern mid-latitudes by roughly a factor of four.

By turning off the biogenic agriculture source in the PRB analyses, the Agency systematically underestimated uncontrollable background in both the NO_2 and ozone reviews. As noted above, the Singh et al. 2007 and Hudman et al. 2007 papers found that lightning was a much larger contributor to NOx species in the upper troposphere over North America than previously thought, by about a factor of four. This also results in a systematic under-prediction of background concentrations and deposition.

The Summary of Chapter 3 requires revision

The summary should document current levels and trends for the relevant species and metrics. As noted above, it does for some species and metrics but not others. It correctly notes that ambient annual NO_2 concentrations have decreased 35 % from 1990-2005 and ambient annual SOx concentrations have decreased 50 % from 1990 to 2005. Figures 1 and 2 document that these trends are part of a longer trend that will continue as highway vehicle and power plant emissions continue to decrease under current control programs. The summary discusses the importance of ammonia from agriculture noting increased emissions over recent decades but does not provide a quantitative estimate of the trend. It summarizes current deposition patterns but not trends.

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[35] ISA at page 3-54.

[36] L. Jaegle, L. Steinberger, R. Martin, and K. Chance, "Global partitioning of NOx sources using satellite observations: Relative roles of fossil fuel combustion, biomass burning, and soil emissions," Faraday Discussions, **130**, 407-423 (2005).

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

Section 4.1 – Introduction to Ecological Concepts

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

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

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

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

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

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

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[37] National Research Council, Valuing Ecosystem Services: Towards Better Environmental Decision-Making, National Academies Press, Washington, DC, 2004, at page ES-6.

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

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

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

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

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

One of the Protocols under the Convention, is the 1999 Gothenburg Protocol to Abate Acidification, Eutrophication and Ground-level Ozone. The Protocol sets emission ceilings for 2010 for four pollutants, sulfur, NOx, VOCs, and ammonia. The ceilings, if fully implemented, would reduce Europe's sulfur emissions by 63 % compared to 1990 levels. For NOx, the ceiling represents 41 % control, and for ammonia 17 % control from 1990 levels. These ceilings were negotiated on the basis of scientific assessments of pollution effects and abatement options. They do not represent the emission reductions required to attain critical loads (or critical levels of ozone) throughout the region, although they are expected to reduce the area of Europe with excessive levels of acidification and eutrophication substantially.

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

[39] ISA at page AX8-2.

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

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

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

Section 4.2 - Ecological Effects of Acidification

Similarities and differences between sulfate and nitrate need to be highlighted

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

[40] J.-P. Hattelingh et al., Status of European critical loads with emphasis on nitrogen, Chapter 1 in Coordination Centre for Effects (CCE) Progress Report 2007,Bilthoven.
[41] R. Skeffington et al., Uncertainty in critical loads assessment models, Science Report SC030172/SR, The Environment Agency, United Kingdom, March 2007.

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

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

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

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

Fourth, trends in N deposition and its potential effects are less clear and the timescale of N saturation may be longer than for S and longer than previously thought. During the 1990s, total N deposition was relatively constant, with NOx emissions decreasing and reduced N emissions increasing over the decade. The ISA notes that during the 1990s the relationship between N deposition and nitrate in surface water was complex with different trends in different areas.[48] Efforts to explain the inconsistent trends were

[42] ISA at page 4-8.
[43] ISA at page 4-13.
[44] ISA at pages AX4-30 and AX5-11.
[45] ISA at page 4-82.
[46] ISA at page 4-40.
[47] ISA at page AX5-11.
[48] ISA at page 4-41.

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

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

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

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

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

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

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

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[49] ISA at page 4-43.
[50] Ibid.
[51] ISA at page 4-22.
[52] ISA at page 4-125.
[53] ISA at page 4-73.

A key question/issue is at what level of a potential indicator do effects appear and what the level of that indicator will be in the future as NOx and SOx reductions already "on the books" are implemented. The ISA should address the first part of this question and the risk assessment should address the second.

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

Section 4.3 – Nitrogen Nutrient Addition

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

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

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

[54] For example, Chapter 3 of the Great Waters Third Report to Congress summarizes numerous national control programs that are reducing NOx emissions and numerous state, local and regional programs that are addressing specific estuaries and their pollution problems around the country. In addition, the Final Report of the Multi-Agency Critical Loads Workshop held in May 2006 summarizes a number of programs underway to apply critical loads in specific regions and ecosystems in the U. S. as a tool to understand and manage sulfur and nitrogen deposition issues.

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

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

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

The discussion of most sensitive and most affected ecosystems in Section 4.3.4 indicates that in the western U. S. there is concern for alpine ecosystems in the Colorado Front Range, chaparral watersheds of the Sierra Nevada, and Southern California sage shrub communities. While there have been high NOx emissions and deposition historically in the Los Angeles Basin and San Joaquin Valley of California, there have been major reductions in NOx emissions over the past several decades and the State Implementation Plans for these areas to meet the current national ozone standard require drastic additional cuts in NOx emissions. Therefore, a national secondary standard is not needed to address NOx effects in these areas of California.

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

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

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

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[55] ISA at page 4-133.
[56] ISA at page 4-149.
[57] ISA at page 4-148.
[58] ISA at page 4-151.

Current deposition of inorganic N from the monitoring sites noted in Chapter 3 is the order of 10 kg N ha^{-1} yr^{-1} or less as shown in Figure 3.8-3. There will be major reductions in NOx emissions from power plants in the eastern U. S. due to the CAIR rule (a 61 % reduction from 2003 levels by 2015 in the CAIR region) and nation-wide from the on-going federal motor vehicle control program throughout the U. S. Therefore, the oxidized N contribution to deposition will decrease substantially in coming years due to controls already on the books.

The implications of the fact that there are many sources that contribute to eutrophication need to be discussed

Eutrophication of estuaries and coastal waters is another major concern related to N nutrient addition. Eutrophication is a condition in which a body of water becomes over-rich in nutrients and results in nuisance or toxic algal blooms and reduced oxygen availability. In the U. S., as described in sections 4.2.3.3 and 4.3, this is a special concern in estuaries and marine ecosystems. Eutrophication is not primarily an air pollution-related problem. It is a matter of both water and air pollution, with N additions from both atmospheric deposition to watersheds, direct deposition to water surfaces, wastewater discharge, agricultural runoff and urban runoff. It therefore involves not only combustion sources of NOx, but also agricultural practices, fertilizer use, treatment of human waste, and treatment of animal waste. The ISA summarizes several studies of the national extent of eutrophication (Bricker et al. 2007) as well as studies of individual watersheds. There is a wide range of findings for the extent of eutrophication and the contribution of atmospheric deposition to specific water bodies. The ISA notes that the estimates of atmospheric contribution entail considerable uncertainty since they depend on many assumptions.[59] The wide range of atmospheric contributions, the wide range of sensitivities of different water resources, and the wide range of contributions from non-atmospheric sources makes using a national standard to address the situation problematic. The ISA should acknowledge that secondary air quality standards cannot, by themselves, solve the nation's eutrophication problems and that the atmospheric contribution of oxidized N to eutrophication is being substantially reduced under current control programs.

There are a wide range of existing regulatory frameworks that are already addressing the eutrophication of estuaries and other waters in the U. S. For example, the Chesapeake Bay Program initiated in 1983 by the EPA and states surrounding the Bay is discussed in the ISA.[60] The Great Waters Report to Congress[61] contains a long list of research and regulatory programs that affect N and other water pollution issues in the country. All these programs at the federal, state, and local levels have been focused on reducing pollutant loads without the existence of separate national secondary standards for SOx and NOx.. Thus, a great deal of regulatory tools and flexibility already exist to allow

[59] ISA at page 4-121.
[60] ISA at page 4-161.
[61] U. S. Environmental Protection Agency, Deposition of Air Pollution to the Great Waters: Third Report to Congress, EPA453/R-05-005, June 2000.

cooperative efforts to study and solve the nation's eutrophication problems. The ISA should acknowledge and enumerate all the different existing regulatory authorities and programs that affect eutrophication.

Chapter 5 – Findings and Conclusions

The chapter is more of a list of findings from the literature than an integrated synthesis of the information that would be the basis for consideration of separate secondary standards. There is no specific discussion of potential indicators although the conclusions section, 5.7, does use the metrics of kg N ha^{-1} yr^{-1} and kg S ha^{-1} yr^{-1}. For example, reference is made to modeling studies of Shenandoah National Park indicating that re-acidification may be prevented if deposition is kept between 9-15 kg N ha^{-1} yr^{-1} and 0-6 kg S ha^{-1} yr^{-1}. In addition, reference is made to several different levels of N deposition that have been associated with effects in different ecosystems and parts of the country.

There is a general statement that "Overall, we conclude that there is a causal relationship between current levels of S and N deposition and numerous biologically adverse effects on ecosystems in the United States." Since the strongest evidence of ecosystem effects comes from locations that have had high historic deposition levels and since deposition levels of S and oxidized N have been decreasing, it is not so clear that current levels of deposition are causing all the adverse effects noted in the ISA. More importantly, since levels of S and NOx deposition will be decreasing substantially over the next several decades under current control programs, it is not clear that additional control of combustion NOx and SOx beyond that which will occur under existing regulatory programs is needed.

There is also a paragraph that indicates that the current regulatory networks for S and N deposition are inadequate to characterize heterogeneity or hotspots. It further states that hotspots have been identified where deposition rates exceed the nearest regulatory monitor by a factor of seven (with no specific cite given). It also states that adverse ecosystem effects are directly linked to hotspots. This is a particularly strange paragraph since it is the conclusions section of the findings and conclusions chapter of the ISA yet there is no mention of N and S deposition hotspots in the balance of the ISA. There is discussion of hotspots in two ways in the ISA and its Annexes. One is in the context of hotspots of denitrification in anoxic sites in wetland soils and the second is discussion of a recent paper that used Hg levels in fish to identify biological Hg hotspots (Evers et al., 2007) which may or may not represent Hg deposition hotspots.

Based on the foregoing general and specific comments, the ISA needs extensive revision before it can form the basis for policy decisions regarding separate secondary standards for SOx and NOx.