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SCIENCE ADVISORY BOARD

December 9, 2010

EPA-CASAC-11-003

The Honorable Lisa P. Jackson
Administrator
U.S. Environmental Protection Agency
1200 Pennsylvania Avenue, NW
Washington, DC 20460

Subject: Review of the *Policy Assessment for the Review of the Secondary National Ambient Air Quality Standard for NOx and SOx: Second Draft*

Dear Administrator Jackson:

The Clean Air Scientific Advisory Committee (CASAC or Committee) Oxides of Nitrogen (NO_x) and Sulfur Oxides (SO_x) Secondary National Ambient Air Quality Standards (NAAQS) Review Panel met on October 6-7, 2010 and held public teleconferences on November 9, 2010 and November 10, 2010 to review EPA's *Policy Assessment for the Review of the Secondary National Ambient Air Quality Standards for NOx and SOx: Second Draft*. This letter provides CASAC's overall comments and evaluation. The CASAC and Panel membership is listed in Enclosure A. The Panel's responses to EPA's charge questions are presented in Enclosure B. Finally, Enclosure C is a compilation of individual panel member comments. This letter provides our views on: 1) the need for retaining the current and developing new secondary standards; 2) limitations in our review of the second draft *Policy Assessment* and priority needs to be addressed in making revisions; and 3) CASAC recommendations regarding the secondary standard.

Need for retaining current and for designing new secondary standards

The current public-welfare-based (secondary) NAAQS standards for oxides of nitrogen (NO_x) and sulfur oxides (SO_x) were designed to protect vegetation from exposures to injurious concentrations of gaseous NO_x and SO_x. This protection is a desirable goal and for that reason the CASAC Panel recommends that the current secondary NO_x and SO_x NAAQS standards should be retained. The current standard for NO_x is an annual arithmetic standard of 53 ppb, using NO_2 as the indicator species, identical to the primary annual health-based standard, with no short-term secondary standard. The current secondary standard for SO_x is 0.5 ppm 3-hour average, not to be exceeded more than once per year, using SO_2 as the indicator species, and it is separate from the primary standard and there is no long-term secondary standard.

EPA staff has demonstrated through the findings of the Integrated Science Assessment (ISA), the Risk and Exposure Assessment (REA), and the draft *Policy Assessment* that ambient NO_x and SO_x can have, and are having, adverse environmental impacts on some ecosystems across the United States due to deposition of NO_x and SO_x, even at current concentrations under existing standards. Those impacts include ecosystem acidification and undesirable levels of nutrient enrichment in some ecosystems. For this reason, CASAC has concluded that different and more protective standards are needed for NO_x and SO_x. A NAAQS is needed that would be formulated in a way that would provide requisite protection in sensitive areas, while not providing protection that is more stringent than necessary.

We compliment EPA Staff on the progress that has been made since CASAC reviewed the first draft *Policy Assessment* and appreciate Staff's responsiveness to CASAC's initial comments. There are significant scientific challenges in developing multipollutant, ecologically relevant secondary standards. These challenges are made more difficult by the regulatory constraints under which EPA staff operates: the standard must use ambient air pollutant concentrations as the indicator, rather than pollutant depositional flux and only oxidized nitrogen, not chemically-reduced forms (both inorganic and organic) of reactive nitrogen, is currently included as a criteria pollutant. To meet these constraints, EPA has developed a new index, the Atmospheric Acidification Protection Index (AAPI). This innovative index integrates the combined effects of atmospherically deposited oxides of nitrogen and sulfur (NO_x and SO_x), as well as chemically reduced forms of reactive nitrogen (NH_x). The AAPI also takes into account a series of ecosystem characteristics that determine sensitivity to total acidifying deposition in various regions of the United States. The AAPI approach is responsive to recent recommendations by the National Research Council for multi-pollutant air quality management (*Air Quality Management in the United States*, 2004).

Despite the regulatory constraints described above, the AAPI approach would appropriately integrate the combined effects of NO_x and SO_x deposition on aquatic acidification, and could provide protection for sensitive aquatic ecosystems at an appropriate spatial scale. Use of the AAPI, however, introduces a number of technical complexities because it considers depositional effects of multiple pollutants within diverse and complex ecological systems. Because of these complexities, it is not apparent how to construct an equally appropriate, and significantly simpler, approach to capture the many important processes that influence the relationship between observable atmospheric concentrations and aquatic acidification.

CASAC notes that EPA did not have sufficient time in preparing the second draft *Policy Assessment* to formulate a complementary approach that would be the basis for standards to protect against nutrient enrichment effects on aquatic ecosystems and nutrient enrichment and/or acidification effects on terrestrial ecosystems. Although a standard that focuses on aquatic acidification would provide some co-benefits in addressing these other adverse effects, in the future, EPA should consider developing approaches for protecting against nutrient enrichment effects on aquatic ecosystems and nutrient enrichment and/or acidification effects on terrestrial ecosystems.

Challenges to the CASAC review of the second draft *Policy Assessment* and priority needs to be addressed in final revisions

CASAC's review of this document has been constrained by the limited time available for review and the incomplete status of the submitted document. Even though the second draft *Policy Assessment* was novel and complex, the CASAC Panel received the document only three weeks before the review meeting. As described below and in our responses to the Charge Questions, there are critical sections of the *Policy Assessment* that are unclear and/or that require further analyses. In addition, and in contrast to policy assessments for other pollutant reviews, EPA did not provide staff recommendations for key elements of the secondary NAAQS for NO_x and SO_x along with supporting rationales. As a result, CASAC is not able to provide specific comments on the EPA Staff recommendations, nor is CASAC able to use Staff recommendations to help frame CASAC discussions or recommendations about the four key elements of the NAAQS.

The final *Policy Assessment* needs to more clearly and fully set out the basis for the recommended ranges of each of the four elements (indicator, averaging time, level and form) of the proposed NAAQS. The implications of choosing specific combinations from within the ranges of elements should be thoroughly discussed, with justifications provided for the specific options or range of options that staff recommends. It would also be useful to see a map and/or tabular estimates of the spatial extent and degree of severity of NAAQS exceedances expected to result from the recommended combinations of the elements of the standard.

The final *Policy Assessment* needs to more fully describe the proposed alternative approaches for landscape categorization of inherent sensitivities to acidification, and how these alternative approaches relate to the different target fractions of water bodies that would be protected at different acid neutralizing capacity thresholds. The implications of choosing specific landscape categorization approaches combined with specific fractions of water bodies to be protected should be thoroughly discussed, with justification provided for the specific combinations or range of combinations that staff recommends.

The final *Policy Assessment* needs to provide a more detailed analysis of the uncertainties associated with the entire AAPI calculation, and of the relative sensitivities of the allowable ambient concentrations to uncertainties in its individual components, including uncertainties introduced by use of models (e.g., CMAQ and the ecological model, MAGIC). The AAPI formulation is unavoidably complex and dependent on critical assumptions and model calculations, which are characterized by various levels of uncertainty. This more complete uncertainty analysis should focus on the overall, cumulative uncertainty estimation including the possible application of Monte Carlo techniques.

CASAC recommendations regarding the secondary standard

While the CASAC Panel is supportive of the AAPI approach developed, CASAC is not able to provide consensus recommendations on all elements of the standard because of the constraints to its review identified above. Further, each element of the standard should be considered in the context of the choices for the other elements. The *Policy Assessment* does not yet provide adequate analyses on all the specifics of a new (and novel) NO_x-SO_x secondary NAAQS. However, CASAC will offer initial comments on some elements of the standard. We agree that a 3-5 year averaging time appears appropriate. CASAC supports the general structure of the AAPI equation and agrees that acid neutralizing capacity is an appropriate ecological measure for reflecting the effects of aquatic acidification. Acid neutralizing capacity targets in the range of 20 to 100 μeq/L appear appropriate to consider at this time.

CASAC recognizes the very tight time lines associated with revising the NO_x and SO_x secondary NAAQS. However, CASAC should have the opportunity to review a more complete *Policy Assessment*, one that provides staff recommendations, the basis for the choices made, the direct supporting analyses for those choices, and the ramifications of alternative choices within the ranges of the alternatives. To this end, we are working through the SAB Staff Office to plan a meeting to review the completed *Policy Assessment* in February 2011.Without this information, we are unable to provide the level of advice that you need and to fill our role under the Clean Air Act. EPA staff must provide complete documents with sufficient time to permit an in-depth review.

Summary

While we have identified various needs for additional analyses and enhanced clarity before the final *Policy Assessment* is published, CASAC remains supportive of the novel approach described in the *Policy Assessment for the Review of the Secondary National Ambient Air Quality Standard for NO_x and SO_x: Second Draft*. We support EPA staff's continuing work on revising the *Policy Assessment* to establish a foundation for a revised NO_x-SO_x secondary NAAQS. This work is groundbreaking and significant for several reasons:

1) The current NAAQS review for welfare-based effects was conducted separately from the review of the health-based standard and has allowed focus on ecological impacts.
2) The review was designed to consider two criteria pollutants at the same time, and set the stage for a "multi-pollutant/multi-media/multi-effect" approach as recommended in the 2004 National Research Council report, and
3) The AAPI takes into account another chemical form of biologically reactive nitrogen, NH_x that is important to aquatic acidification, but is not a criteria pollutant.

In closing, CASAC trusts that our comments will be useful in revising the *Policy Assessment*.

Sincerely,

/Signed/ ***/Signed/***

Dr. Armistead (Ted) Russell, Chair
CASAC Oxides of Nitrogen and Sulfur Oxides
Secondary NAAQS Review Panel

Dr. Jonathan M. Samet, Chair
Clean Air Scientific Advisory Committee

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Enclosure A – Rosters
U.S. Environmental Protection Agency
Clean Air Scientific Advisory Committee
Oxides of Nitrogen (NOx) and Sulfur Oxides (SOx) Secondary Review Panel

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* Provided individual comments only. Did not participate in the Panel's deliberations.

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Enclosure B
CASAC Oxides of Nitrogen (NOx) and Sulfur Oxides (SOx) Secondary Review Panel Consensus Responses to Charge Questions

Chapter 3: Considerations of Adversity to Public Welfare

1. *What are the Panel's views on the definitions of adversity that are appropriate to consider in determining what constitutes adversity to public welfare relative to the NOx and SOx secondary standards?*

Ecosystem services provide a framework to characterize and describe how changes in ecosystem structure and function affect public welfare, even if they cannot be specifically quantified. The link is well-documented between the selected ecosystem effects indicator, acid neutralizing capacity (ANC), and the public welfare effects of lost value of recreational fishing, biodiversity, and habitat. Fish populations (and in some cases whole species) become unsustainable in lakes and streams with decreases in ANC levels caused by elevated inputs of acidic deposition. The text mentions non-use values several times, but it would be helpful to make explicit that this includes values for the preservation of habitat and biodiversity that are independent of human use value. These services generally fall into the category of cultural services. More could be done to explain and characterize the qualitative links between acidic deposition and lost ecosystem services that are known and documented but cannot be specifically quantified for a specific amount of acidic deposition. While it is clear that the total value of these services is large; what is important to convey is the degree to which they are diminished at current acidic deposition levels.

Evidence of community, local and state actions to decrease acidification is informative regarding adversity even though such evidence doesn't provide specific estimates of welfare changes. Also including federal actions, such as the Title IV (Clean Air Act Amendments of 1990) program, to address acidification would be appropriate here.

Chapter 4: Addressing the Adequacy of the Current Standards

2. *What are the Panel's views on staff's approach to translating the available evidence and risk information and other relevant information into the basis for reaching conclusions on the adequacy of the current standards and on alternative standards for consideration? a) In light of the Panel's views on the appropriate definitions of adversity to public welfare (see Chapter 3), do you agree that the current levels of NOy and SOx deposition are adverse to public welfare?*

Yes, the Panel agrees that current amounts of NO_y and SO_x deposition are adverse to public welfare especially with regard to effects on aquatic ecosystems in acid-sensitive regions in various parts of the United States. The Panel also agrees with EPA's historical interpretation that air-pollution-induced effects on ecosystems should be considered “adverse to public welfare” whenever these effects include “disruptions in ecosystem structure and function” that are considered important to the people of this country.

3. *Has staff appropriately applied this approach in reviewing the adequacy of the current standards and potential alternative standards?*

Yes, the panel finds that EPA staff has appropriately reviewed the adequacy of the current standards and potential alternative standards. The current NO_x and SO_x standards were designed to protect vegetation against exposures to SO_x and NO_x. Thus, the current standards address only a fraction of the total nitrogen and sulfur compounds that are causing adverse effects on aquatic ecosystems, and these standards are not designed to protect ecosystems from acidic deposition. None of the elements of the current NAAQS standards – indicator, form, averaging time, and level – are suitable for addressing the long-term (multi-annual) cumulative acidification effects of total atmospheric loads of total reactive nitrogen and sulfur on aquatic ecosystems.

The ISA and REA for the current review (as summarized in Chapters 2 and 3) make it clear that current ambient concentrations of airborne nitrogen and sulfur compounds (including not only NO_y and SO_x, as asked in Charge Question 2 but also ambient NH_x as well as organic forms of nitrogen) are now causing significant “disruptions in the structure and function of aquatic ecosystems” in various acid-sensitive regions of the United States.

4. *Has staff appropriately acknowledged the potential beneficial effects of nitrogen inputs into nutrient limited ecosystems, while maintaining the focus of the review on preventing adverse effects in nitrogen sensitive ecosystems?*

While the potential beneficial effects of nitrogen inputs into nutrient-limited ecosystems have been acknowledged, the tone and emphasis given has not been appropriately balanced. As an example, the last few lines of page 4-45 in Chapter 4 and especially the first four words, may suggest the potential benefits to be very limited: “In certain limited situations, additions of nitrogen can (word inserted) increase rates of growth, and these increases can have short-term benefits in certain managed ecosystems....”

A better balanced presentation of these same ideas could read as follows:

> “Most ecosystems in the United States are nitrogen-limited, so regional decreases in emissions and deposition of airborne nitrogen compounds will lead to some decrease in growth of the vegetation that surrounds the targeted aquatic system. Whether these changes in plant growth are seen as beneficial or adverse will depend on the circumstances. Increased carbon sequestration due to increased growth in N-limited ecosystems may be the most significant category of potential beneficial effects of N deposition.”

Carbon sequestration is not addressed in the *Policy Assessment*. Carbon sequestration is implied, however, by the inclusion of climate-related issues in Table 3-1 on page 3-11. As indicated above, increased carbon sequestration due to increased growth in nitrogen-limited ecosystems may be the most significant category of potential beneficial effects of nitrogen deposition.

While the *Policy Assessment* and supporting documents acknowledge the possibilities of beneficial effects, they tend to minimize them. The panel believes that while such unintended effects by no means justify continuing current levels of air pollution, a balanced document should discuss these unintended effects more thoroughly.

Chapter 5: Conceptual Design of an Ecologically Relevant Multi-pollutant Standard

5. What are the Panel's views on staff's revised conceptual framework for the structure of a multipollutant, ecologically relevant standard for NOx and SOx? To what extent does the Panel agree that this suggested structure adequately represents the scientific linkages between ecological responses, water chemistry, atmospheric deposition, and ambient NOx and SOx?

With some exceptions noted below, the revised conceptual framework and structure of the proposed standard(s) are well-thought out for addressing various components and connections between these components (ecological effects, atmospheric wet and dry deposition, atmospheric concentrations of NO_y and SO_x, and surface-water chemistry).

For example, the framework and the structure "take into account" the reduced ambient NH_x and its deposition in designing the AAPI (atmospheric acidification potential index). The revised treatment of ammonia and deposition of reduced nitrogen is an improvement over the first draft in that AAPI will reflect periodic changes in NH_x concentrations. Emissions of ammonia (which is currently an unregulated air pollutant) and resulting ammonia and ammonium concentrations and deposition amounts are expected to increase over the next few decades because of increased food production and increased activity in CAFO sources (confined animal feeding operations) in the United States.

The conceptual framework for the proposed multipollutant ecologically relevant standard for NO_x and SO_x is sound with considerable support from the scientific literature on how the generation of strong mobile acids results in the acidification of soils and water. Some of the information, however, is not correct or is incomplete. For example, the discussion of sources of nitrate during snowmelt is incorrect in that it suggests that most of the nitrate released is of atmospheric origin. However, most nitrate mobilized during snowmelt is derived from nitrification in the soil itself. Also the assumptions associated with atmospheric sulfur input being equal to drainage water losses are not correct. For example, the soil can serve as a substantial source or sink of sulfur depending upon soil properties and history of atmospheric sulfur sources.

Even though the AAPI conceptual framework is sound and useful in principle, further evaluation of robustness is required. One way to evaluate robustness of the AAPI framework is by using sensitivity and/or uncertainty analysis, as discussed in our responses to Charge Questions 14 and 21. The AAPI can also be tested by the use of time series chemistry data. Where data are available, one could use the AAPI to estimate a functional relationship between AAPI and changes in SO_x and NO_y concentrations. The values of other components of the AAPI (Q, N_{eco}, $[BC]_o$, LNH_x, TNO_y and TSO_x) have already been estimated by EPA or can be determined from measured values. The recommended analysis of historical data should be carried out at more than one location. The changes in predicted AAPI should more or less match the changes in ANC (perhaps with some lag).

Notwithstanding these concerns, the proposed structure adequately represents the scientific linkages between ecological effects, surface water chemistry, atmospheric deposition, and ambient levels of NO_y, SO_x, and NH_x.

6. What are the Panel's views on the appropriateness of considering a single national population of waterbodies in establishing standards to protect against aquatic acidification? What are the Panel's views on consideration of alternative subdivisions of the U.S. to identify the spatial boundaries of populations of waterbodies and acid-sensitivity categories, specifically:

a) the use of Ecoregion III areas to aggregate waterbodies ?

b) the use of ANC to further aggregate Ecoregion III areas into different categories of sensitivity?

c) the relative appropriateness of the suggested methods for categorizing spatial boundaries of sensitivity, e.g. one nation, binary sensitive/less-sensitive classes, cluster-analysis based sensitivity classes, and individual ecoregions?

The justification, logic, and necessity of the spatial grouping classifications were not clear to the panel. The ecoregions approach has conceptual appeal, but the rationale and limitations for classification and aggregation methods must be better articulated for all options described in the *Policy Assessment* before the CASAC Panel can provide meaningful advice.

The first approach (option 1), which considers the whole United States as one unit, has the advantage that it provides for a single deposition metric and is simple and easy to use. However, the single-region approach also has many weaknesses (e.g., over protection for the least sensitive areas and under protection for areas that are most sensitive necessitates having a system with higher spatial resolution) and is probably not a desirable approach. Nonetheless, the panel finds it useful to include discussion of this option for the overall context. On the other hand, the option 2d, which includes 85 ecoregions, may provide an unnecessary amount of complexity, but future analyses could provide support for such a choice. The use of clustering is also conceptually appealing, although the optimal number of sensitivity categories and the degrees of protection that would be provided under the different sensitivity categories are not clear. It does appear to strike a more reasonable balance between oversimplification and unnecessary complexity. The use of ANC appears to be a reasonable basis for grouping ecoregions into a relatively small number of categories, each containing surface waters with similar inherent sensitivities to acidification. This approach is consistent with the overall goal of developing an ecologically relevant secondary standard to protect sensitive surface waters from further acidification and decrease acidifying deposition to levels that will allow those water bodies (that have been deleteriously impacted by acidic deposition) to recover as indicated by increasing ANC values.

The Panel recommends that the final *Policy Assessment* include a more detailed description of the clustering approach and other options, along with clear illustrations of the advantages and disadvantages of the recommended options.

7. What are the Panel's views on the appropriateness of the critical loads that form the basis for the population assessment to determine deposition metrics?

Using the concept of critical loads is logical and appropriate for development of a secondary standard for the biological effects of NO_y SO_x and NH_x. This approach links concentrations of the atmospheric oxidized forms of nitrogen and sulfur with N & S deposition and their acidifying effects on aquatic ecosystems and includes consideration of chemically reduced forms of atmospheric N.

a) What are the views of the Panel on the appropriateness of generalizing the f-factor approach to apply to lakes and streams in the Western U.S. and other portions of the Eastern U.S.

The f-factor approach is a reasonable initial approach to evaluate the response of aquatic ecosystems to changes in atmospheric deposition. However the f-factor approach is based on steady-state calculation but ecosystems are simply not at steady state. Ultimately, it would be useful to apply dynamic models as management tools to evaluate effects of atmospheric deposition on non-steady state ecosystems.

Differences between the use of MAGIC and the SSWC methods to determine background concentrations of base cations are not adequately described in the *Policy Assessment.* The proposed procedures and differences between the two approaches need to be described more clearly

b) What are the views of the Panel on the filtering criteria used to remove lakes and streams that are naturally acidic or not sensitive to atmospheric deposition?

It is justifiable to exclude in advance water bodies impacted by mine drainage. It is not clear, however, why water bodies with low background ANC and high concentrations of naturally occurring organic acid are, likewise, excluded from further consideration since these are often very sensitive water bodies. The rationale for this approach needs to be better explained with examples given, with some discussion of the implications for eliminating these water bodies. The panel needs this information before it can fully and meaningfully respond to this charge question.

8. *What are the Panel's views on the suggested methods for determining appropriate values of reduced nitrogen deposition in establishing NO_x/SO_x tradeoff curves?*

The proposed approach is reasonable and utilizes the available knowledge on levels and distribution of reduced N based on the CMAQ outputs. Potentially the NADP chemistry and PRISM precipitation results could also be utilized. Due to the high deposition velocity of NH_3, steep concentration gradients near the NH_3 source areas can be expected. Therefore averaging N_{red} concentrations over larger areas may lead to missing smaller areas where NH_3 concentrations may be elevated with potentially high ecological effects. Consequently, option "2" is preferable since it allows for additional spatial refinement of sensitive areas to reflect the spatial and temporal heterogeneity of NH_x deposition. A better understanding of spatial and temporal distribution of reduced N, especially NH_3, in the United States is critical. Realizing that estimates of chemically reduced N deposition are viewed as highly uncertain, efforts should be continued to assure the nationwide monitoring of N_{red}, especially in remote areas.

9. *What are the Panel's views on the revised characterization of the deposition transference ratios (TNOy and TSOx)?*

A major concern with T_{NO_y} and T_{SO_x} is that although they are the critical links between NO_y and SO_x ambient concentrations and their deposition, they are derived using a model that has not been thoroughly evaluated for its ability to accurately simulate N and S deposition because of lack of measurements of the required concentration and deposition components. It is recommended that EPA evaluate the stability of these ratios using different models, emissions and meteorological conditions. It is recommended to calculate these ratios for the following model simulations (in addition to what has already been done):

- CMAQ and CAMx models (it is acceptable, in fact preferable, to use different emissions and meteorological conditions)
- Different model grid resolutions (36-km v/s 12-km or even 4-km, if available)

The CMAQ T_{SOx} calculation could also be evaluated using a combination of measured wet deposition data from NADP and the measured concentrations and estimated dry deposition of SO_2 and pSO_4 from CASTNET.

The draft *Policy Assessment* notes the possibility of significant amounts of sulfur and nitrogen deposition in the forest ecosystems in the coarser particle mode and further notes that CMAQ may not adequately account for coarse particle sulfate deposition. At the same time, most currently available measurement programs do not specifically quantify coarse mode sulfate or nitrate concentrations or deposition, so there are only very scant measurement data with which to evaluate related CMAQ estimates. It is not clear how important coarse particle depositoni is and how it should/would be addressed. The Panel requests more clarification on this issue.

On a related topic, the Panel suggests that the Agency consider the feasibility of calculating an alternative deposition transfer ratio for oxidized nitrogen, using a combination of (or perhaps the sum of) nitric acid and particulate nitrate, as an alternative to using NO_y. A possible advantage of this approach is that nitric acid is the component of NO_y that deposits most efficiently, and correlates best with total oxidized N deposition. Consequently, the resulting total deposition estimate would be less dependent on CMAQ model performance. A second possible advantage is that this calculation (as well as the T_{sox} calculation) could be made using currently available and relatively low-cost CASTNET filter pack measurements, without a need to establish a large new network of continuous NO_y and SO_2. A disadvantage of this approach is that while CASTNET measurements of total (gas + particle) nitrate are considered reliable, the CASTNET measurements of the separate HNO_3 and p-NO_3 components are subject to large sampling artifacts.

Estimates of total oxidized N deposition calculated using the original T_{NOY} method and the suggested alternative approach could be evaluated against both CMAQ estimates of total deposition as well as wet deposition measurements from the NADP plus dry deposition estimates from the CASTNETnetwork. It would also be important to consider whether the alternative approach would perform as well as the original T_{NOY} when calculated over broad spatial scales, and over long time periods when NO_x emissions and NO_Y species compositions may change.

As an alternative approach, EPA should attempt to further evaluate the stability of the T_{SOx} and T_{SOx} ratios over time and space recognizing that these ratios are a function of both air concentrations and deposition velocities. One possibility would be to use information from other sources (e.g., CASTNET) to make some comparisons among air concentrations for these chemical species with respect to their modeled deposition velocities and resultant estimated deposition where such data are available. The Panel recognizes that the suite of chemical species that can be used in this analysis is less extensive than that modeled in CMAQ. If these ratios obtained from other data sources show substantial variation over time or space, it would be useful to evaluate the relationship between meteorological and/or emissions sources.

10. What are the Panel's views on staff's conclusion that an averaging time of 3 to 5 years is appropriate given the AAPI form of the standard?

The EPA staff makes a good case for using the averaging time of three to five years and the panel agrees with that recommendation.

11. What are the Panel's views on the preliminary staff conclusions regarding alternative target ANC levels that are appropriate for consideration and the rationale upon which those conclusions are based?

Based on the available scientific data, the range of target ANC values considered in the *Policy Assessment* is appropriate, i.e., 0, 20, 50 and 100 μeq/L as target levels. These values encompass the range of sensitive ANC classes for surface waters in the literature, and there is a range of biological responses corresponding to this range of ANC levels. There will likely be biological effects of acidification at higher ANC values within this range, and there are relatively insensitive organisms that are not impacted at ANC values at the low end of this range. Adverse effects of acidification on aquatic biota are fairly certain at the low end of this range of ANC and incremental benefits of shifting waters to higher ANC become more uncertain at higher ANC levels. There is substantial confidence that there are adverse effects at ANC levels below 20 μeq/L, and reasonable confidence that there are adverse effects below 50 μeq/L. Levels of 50 μeq/L and higher would provide additional protection, but the Panel has less confidence in the significance of the incremental benefits as the level increases above 50 μeq/L. As indicated in the draft *Policy Assessment*, there are clear and marked biological effects at ANC values near 0 μeq/L, so this is probably not an appropriate target value for the AAPI. At a target value of 20 μeq/L, aquatic biota experience acidification effects. Moreover, at this level of ANC many surface waters experience episodic acidification and associated biological effects. As a result, target ANC values of 20 to 100 μeq/L are in the range of appropriate values, while recognizing that there is additional protection at 50 to 100 μeq/L.

a) In light of the Panel's views on the appropriate definitions of adversity to public welfare (see Chapter 3), what are the Panel's views on the appropriateness of the information related to adversity considered by staff in evaluating alternative target ANC levels?

The information on adversity to public welfare associated with the effects of aquatic organisms and ecosystems at different levels of ANC is appropriate given the available literature. The reduction in or loss of sensitive species that would otherwise have been present in that ecosystem is an appropriate pointer to adversity to public welfare. There is relatively little information on the temporal biological response of acid-impacted aquatic ecosystems to marked decreases in acidic deposition. Most of the information on biological response to acidification is developed from spatial data. It may be useful to emphasize that it is unclear if the biological patterns observed for spatial data of varying ANC will similarly occur temporally in surface waters following increases in ANC due to any future decreases in nitrogen and sulfur deposition.

12. What are the Panel's views on the approaches considered by staff for assessing alternative target percentages of water bodies for protection at alternative ANC levels?

This question is difficult to address without specifying the filtering criteria for the watersheds at specific ANC thresholds. As noted in our response to Charge Question 7b, the rationale for the filtering criteria should be better explained. It would be helpful to see an analysis of the implications of different choices of the filtering criteria for the target percentages. It is difficult to suggest target percentages without more information on subdivisions of the United States to be used and the distribution of ANC values in these subdivisions. Since effects at current deposition levels are adverse, the target should be a higher percentage than is currently adversely affected in sensitive areas.

The DL factors, which clearly are numerical indices of some kind, should either be formally defined in the form of equations or it should be made clear how the numerical values for them presented in Tables 5-12 and 5-13 were derived.

Chapter 6: Co-protection for Other Effects Using Standards to Protect Against Aquatic Acidification

13. What are the Panel's views on the utility of the additional analyses of co-protection benefits to inform consideration of alternative levels of the standard?

The analyses and conclusions in Chapter 6 are important because the decision to focus on the effects of acidification on aquatic ecosystems means that in the current standard setting process, other important effects on ecosystems (documented in the ISA), are not being explicitly taken into account. To the extent that standards set to protect against effects of acidification on aquatic ecosystems result in decreased amounts of nitrogen and sulfur deposition there may be additional beneficial and detrimental effects to other ecosystems. It is important to acknowledge these even if they are not quantified.

The analyses reported in Chapter 6 are adequate for this purpose, but the interpretation of the conclusions could be broadened. One analysis suggests that sensitive terrestrial systems located in the same watersheds with sensitive aquatic systems would be protected by the deposition levels that would be needed to protect the aquatic systems. A relevant question then is the extent to which sensitive terrestrial ecosystems are co-located with sensitive aquatic systems throughout the country.

Similarly, even though the standard would not decrease N deposition to the extent required to meet the target share of the TMDL in the Chesapeake watershed, the discussion could say more about what percentage of the target TMDL might be achieved.

The discussion in this chapter should acknowledge that the level of protection from undesirable effects of N deposition in terrestrial ecosystems is not addressed in this analysis and remains uncertain, especially in the arid and semi-arid ecosystems of the Southwest. Negative effects of N deposition on lichen communities are observed in some locations at very low amounts of N deposition.

Introduction of mobile sulfate or nitrate anions into acidic soils (whether naturally acid or acidified by pollution) can result in near instantaneous acidification of waters, whereas acidification of soils is a long-term process occurring over decades or longer. Similarly, recovery of surface waters from acidification could happen relatively quickly if mobile sulfate and nitrate are removed, but recovery of acidic soils is highly questionable as soils in humid systems naturally acidify but do not spontaneously become less acid. The rate of acidification of soils should decrease with decreased atmospheric inputs of sulfur and nitrogen, however.

Chapter 7: Evaluation of Uncertainty and Variability in the Context of an AAPI standard, including Model Evaluation, Sensitivity Analyses, and Assessment of Information Gaps

14. What are the Panel's views on the following:

a. The degree to which the chapter appropriately characterizes the potential role of information on uncertainty, sensitivity, and variability in informing the standards?

The discussion of uncertainty and sensitivity analysis is much improved compared to the first draft in consolidating the uncertainty, sensitivity and variability in the proposed indices; however, a more complete quantitative analysis of uncertainty is needed for the AAPI. The reason it is particularly important to conduct a comprehensive uncertainty and sensitivity analysis for the AAPI form of the standard is that many of the components of the AAPI cannot be evaluated because of lack of measurements. One way to gain confidence in using the AAPI is to examine quantitatively how sensitive the SO_x and NO_y response surfaces are to different components of the AAPI. The Panel's major comments on this chapter include:

1. Summarize the general framework applied for uncertainty analysis, e.g. the World Health Organization framework used in other NAAQS assessments.
2. Extend the uncertainty analysis beyond the components and examine the propagation of the uncertainties through the entire AAPI, with particular focus on how uncertainties impact the levels of NO_2 and SO_2. Include constraints from available observations.
3. Aggressively pursue the identification and reduction of biases in the CMAQ model that are relevant to the AAPI.

It is recognized that it is difficult to quantify the uncertainties associated with the AAPI. Nevertheless, the Panel recommends that EPA pursue more complete uncertainty analysis focusing on the overall, end-to-end, cumulative estimation, including the possible application of Monte Carlo techniques. EPA may want to assess the report presented by the Electrical Power Research Institute at the October 6-7, 2010 meeting.

While there is significant uncertainty associated with model calculations both in CMAQ and the MAGIC/SSWC, there is a considerable amount of empirical observations that provide constraints on the magnitude of these uncertainties. The combined use of uncertainty propagation and the observational constraints should be pursued.

b. The appropriateness and completeness of the evaluation of CMAQ model performance and sensitivity to critical inputs?

The inclusion of comparisons of CMAQ and CASTNET results and the related discussion on the CMAQ limitations in Chapter 7 is very helpful. It should be useful for future improvements of CMAQ. As indicated, the "sensitivity of CMAQ derived deposition transformation ratios to changes in emissions and treatment of chemistry" is not yet completed. This should be a high priority for EPA.

The performance evaluation of the CMAQ model is incomplete. A more complete evaluation with measurements is key to improving confidence in the calculations of the AAPI. The overestimation of SO_2 is a significant systematic error that may lead to a bias and may have a major impact on the estimated deposition and the AAPI overall. A figure similar to Fig 7-5 for CMAQ-CASTNET comparison for SO_2 could be very revealing because the model cannot be directly evaluated for dry deposition due to the lack of measurements, and evaluation of the ability of the model to represent ambient levels can serve as a proxy for its ability to represent dry deposition. It is recommended that following evaluations (using daily or weekly averaged quantities, and showing mean normalized statistics as well as normalized mean statistics) be performed to assess the model performance:

1. Model performance for simulating nitric oxide, nitrogen dioxide, sulfur dioxide, nitrate, ammonium and aerosol nitrate, ammonium, and sulfate for different networks for which the data are routinely available,
2. Model performance for capturing observed levels of wet deposition of sulfate, nitrate, and ammonium using the National Atmospheric Deposition Program (NADP) network
3. Model evaluation using the continuous measurements of nitric oxide, nitrogen dioxide, nitric acid and NO_Y from the SEARCH network in the southeastern United States.

EPA should use the results from the model performance evaluation to describe the uncertainties and limitations of CMAQ simulation of total deposition of reactive nitrogen and sulfur oxides more completely,

c. The utility of the analyses of temporal and spatial variability in the deposition transference ratios (TNOy and TSOx)?

The figures in the *Policy Assessment* showing the spatial pattern of T_{NOy} and T_{SOx} are insufficient to provide the reader with adequate understanding of the spatial variation of the transfer ratios and how they are linked to acid-sensitive ecosystems. The meaning and implications of the box-and-whisker plots are not obvious. The terms “stiff” and “stiffness” are not explained. The T_{NOy} and T_{SOx} are critically important to the APPI calculations, are entirely dependent on CMAQ simulations, and are impossible to fully evaluate with currently available measurements. It is therefore important to demonstrate that their spatial patterns appear reasonable, that the resultant deposition calculations are consistent with (limited) available measurements, and that these ratios remain stable as emissions, concentrations and deposition rates are changed over time.

15. What are the Panel’s views on the insights provided by the AAPI sensitivity analysis including:

a. The evaluation of elasticities of response?

b. The multivariable ANOVA analysis?

Evaluation of elasticity of response is a good way to get an initial estimate of the AAPI sensitivity to its components. However, the Panel recommends doing this analysis also for the SOx and NOx response surfaces to meet a particular standard, as those are the quantities for which compliance with the standard would be determined. The sensitivity analyses should include a larger range of scenarios, such as the sensitivity to the 40% SO2 over-estimation in CMAQ.

A summary is needed for the relative sensitivities of the various parameters that make up the AAPI to show the parameters of the AAPI that have the most and least impact and their influence on confidence levels. e.g., the role of non-atmospheric inputs, including base cation weathering and runoff rates. Such information should be used in driving research and monitoring efforts by EPA.

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16. *What are the Panel's views on the discussion of uncertainty in the critical loads models including MAGIC and SSWC?*

There is clearly uncertainty associated with model calculations. However, the *Policy Assessment* does not acknowledge that there is a considerable amount of empirical field data to support application of this secondary standard. Through monitoring studies there are about 30 years of observations providing a quantitative understanding of the nature and extent of soil and surface water responses to decreases in atmospheric deposition. Through these observations and some field-based experiments, there is also a good understanding of the compensatory response of ANC to changes in concentrations of sulfate and nitrate. These empirical data should be used to evaluate the quality of the AAPI calculations and to support the justification and target parameter values for the AAPI.

There have been limited uncertainty analyses of the ecological models MAGIC and the SSWC. Some uncertainty analysis for MAGIC is presented in the REA. The panel encourages EPA to continue uncertainty analysis of these ecosystem models in the future

Beyond uncertainty analysis, efforts should also be made evaluate model structure and compare this to the structure of other models available for use. Efforts should be made to

- test models, although it is difficult to test steady state models.
- improve and test the N_{eco} calculation by using observed data and improved model simulations of N-deposition.
- compare results from steady-state with dynamic models to obtain a sense for the time scale to achieve target ANC values.
- evaluate the effects of variation and changes in climate on model calculations.

Some of these evaluations may be feasible within the current NAAQS review cycle, while others will help to refine the standards in future reviews.

17. *What are the Panel's views on the areas for future research and data collection outlined in this chapter, on relative priorities for research in these areas, and on any other areas that ought to be identified?*

The future research areas outlined in Chapter 7 are appropriate However, there are other areas that should be considered for future research and data collection.

- Key uncertainties identified in the qualitative uncertainty analysis of Chapter 7 including pre-industrial sulfate, nitrate, base cation, and ANC levels, dry deposition, and ecological indicators, with particular emphasis on parameters that were found to be most influential from the analyses conducted as part of this NAAQS review.
- There is a need to improve understanding of the sources, atmospheric dynamics ambient concentrations, bi-directional transport and deposition of both chemically reduced and organic forms of nitrogen.
- Efforts should be made to develop dynamic models to simulate effects of acidic deposition on soil, drainage waters and biota, to test these models and to apply these as tools in determining critical loads. Research should be conducted comparing results from steady-state and dynamic models.
- There is a need for research to improve the linkages between atmospheric chemistry and transport models with watershed models. Atmospheric models typically have relatively large spatial scales and simulate over relatively short temporal scales. Watershed models simulating acidification of soil and surface waters, in contrast, have small spatial scales and simulate processes over long temporal scales. It is important to quantify the subgrid scale variability in atmospheric deposition and how this variability can be addressed in simulations of watershed response to changes in atmospheric deposition.
- It is essential that surface water monitoring programs be maintained and soil and biological monitoring programs be strengthened.
- There needs to be improvements of tools and models to predict nitrogen retention in watersheds.
- There is a need to better understand the compensatory response of naturally occurring organic acids to decreases in acidic deposition.
- Since the current assessment was unable to address endpoints other than aquatic acidification, there is a need for research regarding endpoints such as terrestrial acidification and aquatic system nutrient enrichment.
- EPA should organize a future workshop to further enumerate and prioritize research identified in the Integrated Science Assessment, Risk and Exposure Assessment, and the Policy Assessment for the secondary NAAQS review for NO_x and SO_x.

The CASAC Panel will discuss priorities among these research and data collection needs at its planned meeting in February 2011.

Chapter 8: Monitoring

18. What are the Panel's views on using an open inlet to capture all particulate size fractions for the purpose of analyzing for sulfate? What is your opinion on using existing CASTNET filter packs as a future Federal reference method for sulfate?

As a prefacing comment on these monitoring questions (18-20), this Panel is pleased to learn that the Agency plans to consult with CASAC to identify the most appropriate monitoring approaches for this NAAQS, and we expect that more informed responses to these and other monitoring questions will be provided in that process. In conducting that monitoring review, we encourage the Agency to emphasize not just compliance determination, but the multiple monitoring objectives outlined in chapter 8 of the *Policy Assessment*, and to consider whether some of those objectives might be most effectively addressed by enhancement of and coordination among existing monitoring programs. In addition, we recommend that the membership of the CASAC monitoring and methods subcommittee be enhanced for that review by adding individuals with expertise in conducting deposition measurements, as well as in assessing the effects of deposited S and N pollutants on aquatic and/or terrestrial ecosystems.

The Panel is not opposed to considering the use of open-faced samplers, and possibly the CASTNET sampler in particular, as a possible federal reference method (FRM) for particulate sulfate, as a component of the multiple pollutant measurements needed to determine compliance with this secondary standard. It should be recognized, however, that the inclusion of coarse particle sulfate (excluded in sulfate measurements by more commonly deployed fine particle samplers) will not by itself provide any information on how much of the sulfate is present in coarse mode particles and which would contribute proportionately more to deposition than their fine particle counterparts. It should also be noted that inclusion of coarse particles, which tend to be alkaline, could lead to formation of positive sampling artifacts from reactions with acidic S and N gasses.

Since the open-faced CASTNET samplers also measure particulate nitrate, and since coarse particle nitrate can contribute to nitrogen deposition, especially in areas influenced by marine aerosols, consideration should be given to evaluating the quality of CASTNET filter pack methods for particulate nitrate as well. CASTNET samplers also measure sulfur dioxide and nitric acid, and so an alternative (to the T_{NOY}) nitrogen deposition transfer ratio could be developed (see response to question 9) based on combined measurements of HNO_3 and pNO_3. As such, all the measurements needed to determine compliance with this standard could be made by the existing CASTNET methods, which could be enhanced by adding new sites in acid and nitrogen sensitive regions, and by adding more detailed measurements like continuous NOy, SO_2, etc. at a subset of those sites to better address important objectives other than compliance. It would be unprecedented to have a compliance network operated by an EPA contractor (as CASTNET currently is) rather than by state agencies. And as indicated above, there are also serious concerns with the quality of CASTNET HNO_3 and particulate nitrate (p-NO_3) data. For these reasons, it would be helpful if the proposed AAMMS review of monitoring methods for implementing this standard includes consideration of both continuous and filter pack measurements of all the relevant S and N species, as well as other approaches like passive samplers and diffusion denuders.

19. *What are the Panel's views on requiring measurements of ammonia and ammonium to assist implementation of the standard?*

Although NH_x deposition estimates could be supplied by CMAQ model output, additional NH_4 and especially NH_3 measurements would be extremely valuable for supporting and implementing the standard both directly - to quantify an unregulated but varying element of the compliance metric - and indirectly, to help evaluate and improve emissions inventories and CMAQ model performance. NH_4 measurements are currently available from the CASTNET and (urban) Chemical Speciation (CSN) networks, and could conceivably be added to the Interagency Monitoring of Protected Visual Environments. NH_3 measurements are currently much sparser and are more critically needed, not only for assessing a key parameter in the AAPI used in the proposed secondary standard but also for better understanding sources and trends of $PM_{2.5}$, regional haze, and sources and effects of N deposition on nutrient enrichment. The passive NH_3 sampling approach currently being deployed in the AMoN Network (at a subset of NADP sites) appears promising and would benefit from more dedicated EPA funding support. Given the current state of development of instruments and methods and their costs, the Panel does not believe that comprehensive ammonia monitoring should be required. The panel strongly encourages, however, that monitoring be conducted at a minimum using the passive NH_3 sampling approach currently being deployed in the AMoN Network.

20. *What are the Panel's views on having a subset (e.g., 3-5 sites) of monitoring stations in different airsheds that measure for the major NO_y species; nitric acid, true NO_2, NO, PAN and p-NO_3?*

Appropriate design of a network required to determine attainment with the proposed standard and to inform future reviews will require a major effort. An appropriate design will be impacted by the choices made in formulating the standard, including the form, indicator, ecoregion approach and fraction of lakes protected. Insufficient information is available at this time, and we commend EPA staff's desire to involve the CASAC monitoring and methods subcommittee in addressing the monitoring related issues. Some initial thoughts are provided below.

As suggested in the response to question 18, a slight modification to the proposed calculation of the deposition transfer ratio (currently expressed as T_{NOY}) for oxidized nitrogen deposition, might allow the use of a modestly expanded version of the existing CASTNET network to determine compliance with the proposed secondary SO_x/NO_x NAAQS. Disadvantages of this approach include the loss of valuable temporal resolution in the weekly aggregated CASTNET filter pack data, uncertainties in the portioning between nitric acid and particulate nitrate, and the exclusion of important NO_y components like NO, NO_2 and PAN, which better reflect the sources of oxidized nitrogen emissions, which eventually contribute to N deposition downwind, and/or which may represent important components of total deposition at some locations.

For these reasons, it is important for implementing this secondary standard that the existing monitoring network be expanded by adding sites in different kinds of sensitive areas, and refined by adding more detailed supplemental measurements at a small subset of these sites. Valuable supplemental measurements would include continuous NO_y and trace SO_2, PAN, true NO_2, and possibly nitric acid, as well as p-NO_3, and NH_3 that would be measured continuously, if practicable, or by integrated methods. Possibly there will be opportunities to add CASTNET filter packs, passive samplers, denuder analyses, and/or other supplemental measurements to several of the existing or planned rural National Core (NCore) Multipollutant Monitoring Network sites. Such measurements would not only help respond to the multiple objectives for this secondary standard outlined on page 8-1 of the PA, they would also be of great value for improving data analysis and modeling assessments of sources, atmospheric chemistry, transport of and the effectiveness of control strategies for ozone, $PM_{2.5}$ and regional haze.

Chapter 9: Conclusions

21. What are the Panel's views on the overall characterization of uncertainty as it relates to the determination of an ecologically-relevant multi-pollutant standard for NOx and SOx?

EPA has done a good job of qualitatively discussing uncertainties in Chapter 7 and reviewing them in Chapter 9. As noted in response to charge questions 14-16, CASAC believes it is important that there is further progress on quantitative analysis of sensitivity and uncertainty, for key components of the AAPI, for the combined effect of multiple uncertainties on the AAPI, and implications for specification of the trade-off between NOy and SOx allowable concentrations. In Chapter 9, EPA should provide a concise summary of those key uncertainties that are most likely to lead to bias, imprecision, or both, in the AAPI, and the implications of such uncertainties when translating an ANC target into an associated AAPI level. For example, given biases, should the selected AAPI be higher or lower than implied by a specific target ANC? Given imprecision, what range of AAPI might be consistent with a particular target ANC? EPA should conduct a more complete evaluation of the CMAQ simulations used to calculate the deposition transfer coefficients and consider additional processes, such as internal sulfur sources, in the AAPI.

The choice of averaging times needs to inform the variability and uncertainty analyses of Chapter 7. This is because the range of variability and uncertainty depend on averaging time. Similarly, the geographic scope needs to be taken into account in the analysis of variability and uncertainty.

22. What are the Panel's views on the following:

a. The insights that can be gained into potential alternative additional secondary standards (using the AAPI form) by considering:

- *i. Information from studies on the relationship between mortality in aquatic organisms and pH and ANC?*
- *ii. Information from studies on the relationship between fish health and/or biodiversity metrics and pH and ANC?*

iii. *Information on the relationship between pH, Al, and ANC?*
iv. *Information on target ANC levels identified by states and regions, as well as other nations?*

Each of the sources of information mentioned in the charge question both separately and taken together provide a compelling case on the relationships between ANC and other water quality metrics that are associated with biotic health of waters, and provide insight regarding target ANC values. Text should be provided on the validity of spatial survey data when applied to infer temporal relationships. Different states and nations have identified different target levels. Some use pH, others use ANC. It will be helpful to explain and compare how these values were developed. Chapter 9 could clearly and briefly summarize possible co-benefits and unintended consequences of various alternatives for the standard. For example, to what extent might a standard focused on aquatic acidification also be protective of terrestrial acidification or aquatic nutrient enrichment? Would higher levels of target ANC provide more protection for these other effects? These points can be made while still placing emphasis on the sufficiency of the scientific evidence supporting the need for a revised standard to protect from aquatic acidification.

b. The appropriate role of qualitative and quantitative characterizations of uncertainty in developing standards using the AAPI form?

Conceptually, the AAPI approach is compelling and appropriate. There are uncertainties associated with the practical use of AAPI that should be more fully evaluated. The sensitivity and uncertainty characterization of the AAPI needs to include not only statistical analyses associated with specific model parameters individually, but also consider their joint effect (taking into account covariance and dependencies) and an evaluation of possible omissions (e.g. reduced nitrogen inputs, contribution of sulfate sources and sinks in soil). To the extent possible, biases and imprecision in values of AAPI associated with a target level of ANC should be quantified, and these uncertainties should be used to inform specification of ranges of AAPI associated with a target ANC that may be more or less protective within the range of scientific uncertainty. This would lead to a family of NO_y-SO_x trade-off curves associated with each target ANC for a given geographic location. A specific standard would be set by choosing an AAPI within the range of scientific uncertainty, which would then be associated with just one NO_y-SO_x trade-off curve per region. EPA staff is encouraged to offer reasonable judgments about the range of uncertainty in AAPI for a given ANC target based on factors difficult to quantify within the time period of the assessment, such as the preindustrial cation weathering, the deposition transfer ratios, unmodeled factors, ancillary benefits, and unintended consequences.

c. The role of considerations regarding the relationship of the standard to:
i. *the time trajectory of response, e.g. when specific ANC levels are likely to be realized given a specific level of the AAPI?*

Based on recent observations and dynamic model calculations, the time response to recovery in ecosystems from decreases in acidic deposition is very slow. Because of accumulation of sulfur in soils, it is likely that the timescale for recovery of watersheds, especially in the Southeast, would likely be even longer. Factors such as changes in climate and carbon dioxide concentration in the atmosphere could affect the time trajectory, and the effects may be substantially different between aquatic and terrestrial ecosystems.

ii. the likelihood of damages to aquatic ecosystems due to episodic acidification events given a specific target for chronic ANC?

Based on surface waters studied in the Northeast, decreases in ANC associated with snowmelt is approximately 50 μeq/L. Thus, based on these studies, a long term ANC target level of 75 μeq/L would generally guard against effects from episodic acidification down to a level of about 25 μeq/L.

iii. the levels of co-protection for terrestrial ecosystems against acidification effects and the for aquatic and terrestrial ecosystems against effects of excess nutrient enrichment?

There may be co-benefits for terrestrial and coastal ecosystems with respect to decreases in methylization of mercury associated with decreases in sulfur dioxide emissions and decreases in bioaccumulation of mercury associated with increases in pH and ANC. Aquatic ecosystems may not be more sensitive to acidic deposition than terrestrial ecosystems. Many soil time series studies suggest ongoing soil acidification while surface waters are recovering from acidic deposition. This may also suggest that soil is more "sensitive" to inputs of acidic deposition than surface waters. Levels of protection provided by the proposed standard against nutritional nitrogen effects in terrestrial ecosystems are uncertain, especially in arid and semi-arid zones, and should be evaluated in the future.

23. What are the Panel's views on Staff's conclusion that the existing secondary standards for NO_x and SO_x should be retained to provide protection against direct adverse effects to vegetation due to gas phase exposures?

Based on the information presented in the PA, the scientific understanding of effects from direct foliar exposures to gaseous sulfur and nitrogen oxides has not changed appreciably, and the existing secondary standards for SO_2 and NO_2 should be retained. The indicators, averaging times, levels and forms of the current standards are not appropriate for addressing the (indirect) effects of SO_x and NO_x deposition to acid-sensitive ecosystems. Therefore, additional secondary standards to protect against adverse effects from acidic deposition should be added to the existing secondary standard.

24. In light of the Panel's views on what constitutes adverse effects to public welfare (see Chapter 3), what are the Panel's views on:

a) the degree to which current levels of NO_y and SO_x deposition are adverse to public welfare based on evidence and risk information, and information on adversity provided in Chapters 2,3, and 4?

Current and cumulative levels of NO_y and SO_x deposition have been shown to result in environmental damage to an extent that is adverse to public welfare. The effects include acidification of aquatic and terrestrial ecosystems and nutrient enrichment. However, the panel believes that descriptive information about that adversity and its significance could be better and more comprehensively articulated, and that additional discussion of the possible benefits of S and N deposition would be helpful.

b) *target values for ANC that protect against adversity to public welfare in light of the information presented in Chapter 5 concerning levels of ANC and the ecosystem effects associated with those target ANC levels?*

There is substantial confidence that there are adverse effects at ANC levels below 20 μeq/L, and reasonable confidence that there are adverse effects below 50 μeq/L. Levels of 50 μeq/L and higher would provide additional protection, but the Panel has less confidence in the significance of the incremental benefits as the level increases above 50 μeq/L.

c) *factors relevant in selecting target percentages of waterbodies to protect at alternative target ANC levels to protect against adverse effects to public welfare, and weights to place on those factors?*

The justification and implications of alternative spatial grouping classifications were not clear to the panel. It is not clear what is gained by the added complexities of going beyond the two groups of sensitive and not sensitive, although there is inherent appeal to taking into account available information about variations across eco-regions. Because there is large variability in inherent sensitivities of water bodies to acidification effects among different regions and even within regions, protecting a target percentage of lakes from the populations which are potentially susceptible to acidification seems logical, however. It seems that the target should be higher than the current percentages of sensitive water bodies that are below the target ANC.

d) *alternative standards for NOx and SOx that would protect against adverse effects to public welfare based on the AAPI form, and taking into account*

- *consideration of target levels of ANC (chapter 5),*

The panel concurred that ANC levels from 20 to 100 were appropriate to consider.

- *target percentage of water bodies to protect (chapter 5),*

The panel believes that this choice was a value judgment and somewhat arbitrary. Insufficient analysis was provided to adequately support a choice at this time. The panel stresses that the target percentage will also be influenced by whether and how the filters used to exclude lakes (e.g., naturally acidified lakes, for example) are applied.

- *consideration of relevant uncertainties in AAPI components (chapter 7),*

The Panel spent considerable time discussing how and what is necessary to characterize relevant uncertainties in AAPI components in order to answer this and other questions about the PA.

The current sensitivity and uncertainty analysis should be strengthened. For suggestions on specific AAPI components see the responses to charge questions 5, 9, 14, 15, 16 and 22. The panel would also particularly like to see some assessment of the cumulative uncertainties associated with the complete AAPI calculation. One approach to this might be to employ available measurement data and model calculations to compare levels and changes in AAPI estimates over the past 20 years with concurrent ANC levels in surface waters. In some cases, individual components of the APPI could also be compared with their measured counterparts over the same recent time period. The goal of these syntheses and analyses would be to lend defensibility to the approach, provide broad bounds on uncertainties, or, in some cases to provide reality checks on the components of the AAPI.

- *any other potentially relevant factors, such as levels of co-protection against terrestrial acidification and nutrient enrichment (chapter 6)?*

It seems likely that a standard that decreases acidifying deposition to acid-sensitive ecosystems would provide some co-protection benefits to acid-sensitive terrestrial ecosystems. If attaining such a standard results in regional-scale reductions in nitrogen deposition, there may also be reductions in plant growth rates in aquatic or terrestrial ecosystems or components of those ecosystems. These growth rate changes might be viewed as either benefits or dis-benefits, depending on the specific ecosystem and management objective. It is not currently possible to provide quantitative estimates of co-protection benefits or detrimental effects, but it would be useful to qualitatively discuss these associated effects in the final *Policy Assessment* document.

Enclosure C
Compilation of Comments from Individual Members of the CASAC Oxides of Nitrogen (NOx) and Sulfur Oxides (SOx) Secondary Review Panel

Comments from Dr. Praveen Amar

Chapter 3: Considerations of Adversity to Public Welfare

Charge Question 1. What are the Panel's views on the definitions of adversity that are appropriate to consider in determining what constitutes adversity to public welfare relative to the NOy and SOx secondary standards

Chapter Three covers three areas of :a) adversity to public welfare, b) application of ecosystem services framework (provisioning, regulating, cultural, and supporting) as a way to address adversity to public welfare, and , c) usefulness of economic valuation approaches to "value"/monetize ecosystem services, when possible. The second draft of the Policy Assessment is a great improvement over the first draft. The definition of adversity used in this document is derived from, and based on, recent applications of the concept by EPA in other recent environmental policy contexts and is quite applicable to ecosystem effects from exposure to ambient levels of SOx and NOy.

The Chapter is much improved in describing the *current* level of ecosystem services as well as *scale* of adversity to public welfare driven by changes to ecosystem services as a function of *changes* in atmospheric deposition of SOx, NOy, and potentially *no changes* (potentially increases) in atmospheric deposition of reduced NHx. The Chapter presents many quantitative estimates in dollars when economic valuation/monetizing were possible. Also, monetized benefits of current status of ecosystem services are clearly presented in many Tables.

Specific Comments on Chapter 3:

It would be useful if public welfare/adversity was more clearly discussed for, and separately allocated to, NOy and NHx (both through atmospheric deposition and through water runoffs) instead of just atmospheric NOy alone. (see page 3-8; TMDL discussion for Chesapeake Bay; this discussion should be more explicit in describing the role of NHx through water discharge and air deposition; please also see Page 3-25, Line 15, nutrient enrichment refers there "only to that due to NOy deposition").

For Figure 3-6, the range for high end of N and S deposition (300- 1,337 eq/ha-yr) for the Western U.S. is too large and needs to be sub divided (say, in two or three parts) for finer representation of high-end deposition levels in the West.

Sections 3.3.2 and 3.3.3 on economics framework and its role in defining adversity are very well written.

The Section 3.3.4 on "collective action as an indicator of public preferences" correctly notes the actions and efforts on the part of communities, NGOs, and States to reduce acidity of lakes and streams. This Section overlooks what I believe is the most important action/effort taken so far in the U.S. by the federal government: Title IV of the 1990 CAAA to lower SO_2 emissions by 10 million tons per year as well as NOx emissions by 2 mm tpy to address ecosystem acidification. The value of the this national "revealed preference" should be valued/monetized at about $5 billion/year, based on $500/ton of SO_2 controlled and should be noted in this section.

Section 3.3.1.1 needs to be written more clearly to make the points it is "trying" to make with references to table 3-2 and 3-3. I found it hard to understand.

Finally, Table 3-6 (page 3-33) needs major improvement in format including column headings.

Chapter 5: Options for Elements of the Standard

Charge question 5: What are the Panel's views on staff's revised conceptual framework for the structure of a multipollutant, ecologically relevant standard for NOx and SOx? To what extent does the Panel agree that this suggested structure adequately represents the scientific linkages between ecological responses, water chemistry, atmospheric deposition, and ambient NOx and SOx?

The revised conceptual framework and structure of the proposed standard (s) is very well-thought out for addressing various components and connections between these components (ecological effects, atmospheric wet and dry deposition, atmospheric concentrations of NOy and SOx, and surface water chemistry), with one major exception noted below. I had made this same point for the first draft of the policy assessment document.

Even though the framework and the structure "takes into account" the reduced ambient NHx and its deposition in designing AAPI (atmospheric acidification potential index) , it does so in a manner such that future control strategies and policy options most probably will not allow EPA to address and require reductions in U.S. ammonia emissions under proposed standard setting structure. Ammonia emissions are currently at about 4 to 5 million tons per year. Emissions of ammonia (which is an unregulated air pollutant) and resulting ammonia and ammonium concentrations and reduced nitrogen deposition levels are only expected to increase by as much as ten percent over the next few decades because of increased food production and increased activity in CAFO sources (confined animal feeding operations) in the U.S.

Notwithstanding my concern about not addressing reduced nitrogen/NHx directly, the proposed structure more than adequately represents the scientific linkages between ecological effects, surface water chemistry, atmospheric deposition, and ambient levels of NOy and SOx.

Charge Question 6: What are the Panel's views on the appropriateness of considering a single national population of waterbodies in establishing standards to protect against aquatic acidification? What are the Panel's views on consideration of alternative subdivisions of the U.S. to identify the spatial boundaries of populations of waterbodies and acid-sensitivity categories, specifically:

a) the use of Ecoregion III areas to aggregate watrebodies?

b) the use of ANC to further aggregate Ecoregion III areas into different categories of sensitivity?
c) The relative appropriateness of the suggested methods for categorizing spatial boundaries of sensitivity, e.g., one nation, binary sensitive/less-sensitive classes, cluster-analysis based on sensitivity classes, and individual ecoregions?

The first approach (option 1) that considers the whole U.S. as one unit and provides for a single deposition metric is simple and easy to calculate, but its weaknesses are too many to consider this as the preferred approach (e.g., over protection for the least sensitive areas and under protection for areas that are most sensitive).

The three sub-options under second option seem to have merits. However, they are based on the concept of "Level 3 Ecoregions," which is rather poorly described in the document and I was not sure how this approach divides US into 120+ acid-sensitive categories. A reference is made to Omernik's (1987, 1995) and other works about "the analysis of the patterns and the composition of biotic and abiotic phenomena that affect or reflect differences in ecosystem quality and integrity….." What is not explained is how "hierarchical levels are developed" at various levels (Level I, II, III, and IV (future)).

Between options 2a, 2b, and 2c, the approach based on cluster analysis (option 2b) seems to provide the right balance when compared to approach that is not detailed enough (option 2a) or detailed too much (option 2c).

Charge Question 9: What are the Panel's views on the revised characterization of the deposition transference ratios (TNOy and T SOx)?

The policy assessment document proposes to use the output of CMAQ model to calculate deposition transference ratios for both NOy and SOx. The CMAQ hourly predictions at the scale of 12-km grid will be averaged to provide annual transference ratios so as to be consistent with depositional loads derived from ecosystem models. It is not clear how to account for wet and dry deposition of those nitrogen and sulfur species (which ones?) that are not explicitly modeled in the CAMQ. The PAD does note the possibility of large amount of sulfur and nitrogen deposition in the forest ecosystems in the coarser particle mode and that CMAQ's simulations do not account for deposition in the coarse particle mode. It is not clear how big this issue is and how it should/would be addressed.

Charge Question 10: What are the Panel's views on staff's conclusion that an averaging time of 3 to 5 years is appropriate given the AAPI form of the standard?

The PAD makes a good case for using the averaging time of three years (Figure 5-22 on the magnitude of coefficient of variation (CV) shows that it is less than 25%, based on CAMQ simulations for the years 2002-2005).

Comments from Dr. Andrzej Bytnerowicz

7. What are the Panel's views on the appropriateness of the critical loads that form the basis for the population assessment to determined deposition metrics?

Using a concept of Critical Loads is logical and appropriate for development of a secondary (welfare) standard for biological effects of NO_x and SO_x. This approach links concentrations of the atmospheric oxidized forms of nitrogen and sulfur with N & S deposition and their acidifying effects on aquatic ecosystems. What is important is also a fact that the proposed approach includes reduced forms of atmospheric N as a contributor to acidification of lakes and streams.

a) What are the views of the Panel on the appropriateness of generalizing the f-factor approach to apply to lakes and streams in the Western U.S. and other portions of the Eastern U.S.

The purpose of the F-factor is to obtain estimates of the pre-industrial surface water base cation concentrations needed for calculation of critical loads. These values can be obtained from the SSWC and MAGIC models.

At this point I am not able to adequately answer the posed question. Explanation of the problem and graphs illustrating differences between the two approaches are not sufficient for understanding the proposed concept.

I believe this question could be modified to: "Is the proposed methodology for obtaining BC_o values adequately described and what is the Panel's opinion on extrapolating the knowledge gained for the Adirondacks lakes and the Southern Appalachian streams to the rest of the US water bodies?"

b) What are the views of the Panel on the filtering criteria used to remove lakes and streams that are naturally acidic or not sensitive to atmospheric deposition?

It's logical to eliminate in advance water bodies impacted by mine drainage, however, advance elimination of water bodies impacted should be considered and the rationale better explained with examples given, preferably in a distribution form. The panel needs this information before it can fully respond to the question. The panel is concerned about removing lakes and streams with high concentrations of organic acids and with low historical ANC from the analysis since these are often highly sensitive water bodies.
8. What are the Panel's views on the suggested methods for determining appropriate values of reduced nitrogen deposition in establishing NO_x/SO_xx tradeoff curves?

The proposed approach makes sense and utilizes the best available knowledge on levels and distribution of reduced N deposition, however, reliance on the CMAQ-derived values provides high level of uncertainty (see my comments below).

Due to a high NH_3 deposition velocity, steep concentration gradients near the NH_3 source areas exist. Therefore averaging N_{red} concentrations over larger areas may lead to missing smaller areas where NH_3 concentrations may be seriously elevated and with potentially high biological and ecological effects. Therefore option 2 "allow for additional spatial refinement of sensitive areas to reflect the heterogeneity of NH_x deposition" seems to be preferable.

As stated in previous CASAC reviews, a better understanding of spatial and temporal distribution of reduced N, especially NH_3, in the US is critical. Efforts should be continued to assure the nation-wide monitoring of NH_3 in remote areas.

Additional remark: It would be good to develop similar methodologies that account for atmospheric organic N.

11. What are the Panel's views on the preliminary staff conclusions regarding alternative target ANC levels that are appropriate for consideration and the rationale upon which those conclusions are based?

Focusing on a range of ANC values between -50 and 50 μeq/L makes sense from a perspective of the expected pH changes, Al toxicity and related biological effects. At values <-50 μeq/L no further damage should occur, while at values > 50 μeq/L no more improvement is expected.

Improved biodiversity of fish populations may continue up to160 μeq/L and therefore the best protection would be achieved at ANC values >100 μeq/L. However, considering that such recommendation could be impractical, the proposed ANC 50 μeq/L as a target value seems to be reasonable and should be supported.

a) In light of the Panel's views on the appropriate definitions of adversity to public welfare (see Chapter 3), what are the Panel's views on the appropriateness of the information related to adversity considered by staff in evaluating alternative target ANC levels?

Appropriate information has been provided for the aquatic ecosystems. However, I would like to see a better discussion of what the main ANC values considered (20, 50 and 100 μeq/L) would mean to the surrounding terrestrial ecosystems in various eco-zones. That could be discussed for such sensitive indicators and sugar maple and red spruce in the eastern part of the country, and for lichen communities in such areas as Sierra Nevada Mountains in the West.

12. What are the Panel's views on the approaches considered by staff for assessing alternative target percentages of water bodies for protection at alternative ANC levels?

This question comes to an issue of toxicity (damage to individual species) versus the biodiversity changes. What should be more important is where or if there is a common denominator for these two approaches? An approach that would provide various levels of protection against toxic effects and biodiversity changes would be most desirable for scientists, managers and decision makers.

Additional general comments

In general I support the proposed CL-based approach for the newly developed secondary standard. However, at the same time I have to emphasize that such an approach could only work if NOy (NOx plus HNO_3, HONO, PAN, particulate NO_3 and other oxidized reactive N species) replaces NOx (mainly NO and NO_2) as the secondary pollution standard. Correlation between ambient concentrations of NOx and dry deposition of oxidized N to watersheds is poor. This is not surprising because while NO_2 and NO dominate atmospheric budget of the oxidized N, their contribution to that deposition is low due to their low deposition velocities to vegetation and soils. It is well established that N dry deposition of oxidized N is dominated by HNO_3 (which is a component of NOy, but not of NOx) characterized by very high deposition velocity. These problems are well covered on page 4-21 and shown on Fig. 4-21 of the reviewed PA.

Another major problem of the proposed approach is its reliance on the modeled concentrations of NOy, NHx and deposition of oxidized & reduced N from the CMAQ model runs. These modeled outputs are characterized by high level of uncertainty and temporal limitation (model runs are just for certain years). Therefore verification of the model results, both for ambient concentrations and estimates of deposition, is critically needed. Additionally, deriving deposition transference ratios (T values) by dividing modeled deposition of oxidized N by a sum of concentrations of dry and wet N oxidized species will very likely result in highly unreliable values. These values will also differ in time and space considering huge regional and year-to-year differences in ratios of gaseous /particulate/water-dissolved oxidized N and ratios of dry/wet deposition. Better understanding of such issues is needed while developing this approach.

Another problem, mentioned many times in the CASAC deliberations, is that while concentrations of NOx and NOy slowly diminish, concentrations of NH_3 from agricultural emissions and from 3-way catalytic converters are increasing as well as the importance of N_{red} in N deposition. In my opinion there a clear need for regulating all those NHx emissions and development of a new federal air pollution standard for NH_3.

Comments from Ms. Lauraine Chestnut

Charge Question 1:

What are the panel's views on the definitions of adversity that are appropriate to consider in determining what constitutes adversity to public welfare relative to the NOx and SOx secondary standards?

Overall, the presentation and explanation of available information on losses in ecosystem services and associated economic valuation as a result of NOx/SOx deposition is much improved in clarity and context from the first draft PA.

The link is clear and well-documented between the selected ecosystem effects indicator, ANC, and the welfare effects of lost value of recreational fishing as fish populations (and in some cases whole species) are not sustained in lakes and streams with lower ANC levels. The available quantitative information is well presented and explained (except for a few specific questions noted below). However, more could be done to explain the qualitative links between deposition and lost ecosystem services that are known and documented, but cannot be specifically quantified for a specific amount of deposition.

For example, on page 3-13, changes in biodiversity, which are listed as an ecosystem effect of deposition, are associated with changes in cultural ecosystem services related to the preservation of natural areas (nonuse values) in addition to productivity, recreational viewing and aesthetics services that are listed currently in the text. It is well-established that there is public welfare value to protection and preservation of natural ecosystems in condition that supports the flora and fauna species that are native to the system, even when there is no direct use value. This is evidenced in the state and federal statutes that set aside parks and wilderness areas (noted in the first sections of chapter 3), and in willingness-to-pay study results such as the Banzhaf et al. (2006) study discussed on page 3-29. The text mentions nonuse value several times, but it would be helpful to make explicit that this includes value for the preservation of habitat and biodiversity independent of human use value.

Specific comments/questions in Chapter 3

page 3-9: What is the pollutant referenced in the critical loads shown for Europe?

page 3-11: Add nonuse to ecosystem services listed for water.

pages 3-14 and 3-15: Figure 3-5 includes federal and state public lands according to the legend, but the text on page 3-14 just references Class I areas, which I think are just federal. Please make clear what areas are included in the maps, and what oher natural areas may not be included that the public may also care about protecting.

page 3-17, line 20: Ecosystem services provide a framework to characterize and describe how changes in ecosystem function affect public welfare, even if they cannot be specifically quantified.

page 3-18: It is important to recognize and include language that preferences are not just about people's own use and enjoyment of an ecosystem, but also include preservation and bequest value.

page 3-18, second paragraph: Good discussion and explanation of how preferences depend on information.

page 3-25: figure 3-7. It would be helpful for more general audiences to include words such as biodiversity and habitat preservation under cultural services. This is part of "nonuse" services, but I'm not sure most people are aware of this.

page 3-26: It might be useful to reference the language on page 3-22 about the goal of keeping the Adirondack Forest Preserve as "wild forest lands" and "kept in natural conditions." This is a significant motivation behind the public's willingness to pay to prevent the effects of deposition in these areas and is part of what makes the effects adverse to public welfare.

page 3-28: I'm a little confused by Table 3-2. Are these threshold categories mutually exclusive?

page 3-29: Are the results in table 3-3 additive? For example, if a threshold of 100 is met, is the annual value of additional recreational fishing services for NY residents the amount in the bottom row only, or the sum of the 3 rows? If they are all based on a comparison to background, then why are the numbers smaller for the 100 threshold than for the 50 threshold? These numbers reflect just a portion of benefits, as noted in the text, so it is important to include more information in the table title and headings about what they are: recreational fishing services for NY residents.

page 3:30: Same question for table 3-4.

page 3-34: Is there some descriptive information from the REA or the ISA to give a sense of the overall magnitude of the red spruce and maple decline attributable to deposition? The estimates of lost commercial forestry value in the second paragraph are interesting, but are these forests significant timber resources? What can be said to describe the implications of the health of these tree species on the natural habitat and health of the natural ecosystems where these species are prominent? It seems like a more comprehensive story could be summed up here about the loss in services that is linked to deposition, even though specific quantitative valuation is not possible. Perhaps more could be said about the Jenkins et al. (2002) results for avoiding a "significant decline in health" of high elevation spruce in the Southern Appalachians. How does the description of decline in this study compare to what has been linked to current deposition levels? The results indicate substantially greater value than was estimated as commercial forest losses.

page 3-36: Need to say something about how these services are hurt or impaired by eutrophication. The total value of these services is only relevant if something can be said about how they are diminished by the effects of deposition. It is okay if this is only descriptive, but the link needs to be made.

pages 3-40 and 3-41: There is better clarity than in the first draft PA when total values of ecosystem services are presented to give context for the potential effects of deposition. The discussion on page 3-41 is helpful in describing why the CSS and MCF ecosystems are important and how the effects of deposition are likely to diminish the services that these systems provide. Anything that could be added about the extent of the current degradation of these ecosystems due to deposition would be helpful for understanding whether the effects of current deposition are adverse to public welfare.

Charge question 4

Has staff appropriately acknowledged the potential beneficial effects of nitrogen inputs into nutrient limited ecosystems, while maintaining the focus of the review on preventing the adverse effects in nitrogen sensitive ecosystems?

It seems to me that the PA is careful now to acknowledge the potential beneficial nutrient effects of N deposition in some systems. This will come up again when it is time for regulatory assessment, because there may be some loss in benefits when N deposition is reduced.

Charge question 13

What are the panel's views on the utility of the additional analyses of co-protection benefits to inform the consideration of alternative levels of the standards?

The analysis and conclusion in Chapter 6 are important because the decision to focus on the effects of acidification on aquatic ecosystems means that in this current standard setting process, other important effects on ecosystems (documented in the ISA), are not being explicitly taken into account. To the extent that standards set to protect against effects of acidification on aquatic ecosystems also provide some amount of protection against the other effects of deposition, then this provides support that the proposed standards are justified and beneficial.

The analyses reported in Chapter 6 seem adequate for this purpose, but the interpretation of the conclusions could perhaps be broadened. It is clear that standards set to protect aquatic resources from adverse effects of acidification would not fully protect against the effects of deposition on acidification of terrestrial resources and nutrient effects on terrestrial and aquatic resources. However, some partial protection that would be provided could be characterized more fully. For example, the analysis suggests that terrestrial systems located in the same watersheds with acid sensitive aquatic systems would be protected by the deposition levels that would be needed to protect the aquatic resources. So, the question that comes to mind is what do we know about where sensitive terrestrial systems are located relative to sensitive aquatic resources throughout the country. Are they mostly located near one another, or do they occur in completely separate locations in significant amounts? Given the regional nature of ambient NOx and SOx concentrations, how close together would sensitive aquatic and terrestrial resources have to be for protections for one to extend to the other?

Related to this is whether there is benefit to reductions in deposition that are short of the targets for full protection. This depends on whether the dose-response relationships are continuous or substantially nonlinear.
Similar questions come up for the analysis of reductions in N deposition relative to the TMDLs for the Chesapeake watershed. The discussion on page 6-6 shows that N deposition could be higher under an ANC target of 50 than would be allowed given the TMDL target. However, this is the maximum that the N deposition could be if SOx deposition were zero. There is a good chance it would be lower than this. Also, how does this N deposition compare to current levels? How much of the reduction to the target TMDL would be achieved?

Charge question 24

In light of the panel's views on what constitutes adverse effects to public welfare, what are the panel's views on:

a) the degree to which current levels of NOy and SOx deposition are adverse to public welfare?

   The case is well made in the PA, based on the information from the REA and the ISA and information added in the PA, that current levels of NOy and SOx deposition are harming sensitive ecosystems to an extent that is adverse to public welfare. A bit more can be done to carry forward the descriptive information about the significance of the current effects that cannot be fully quantified so that the implications for adversity to public welfare are more comprehensive.

b) target levels of ANC that protect against adversity to public welfare?

   The case seems well supported for a target ANC of at least 50. The wording used to describe the benefits of a target higher than 50 seems unnecessarily cautious. What I understand is that at 50, most sensitive species would survive, but not necessarily thrive. It is certainly clear that loss of an entire species of fish that would otherwise be expected to live in such waters is an adverse effect, so a target of 50 to prevent loss of species is justified. To the extent that the size and robustness of the populations matter to public welfare (and I think there is evidence that they do) then it seems there would be further benefits of an ANC target higher than 50. It may be difficult to quantify the value of this additional benefit, but is it really all that uncertain that there would be some additional benefit?

c) factors relevant in selecting target percentages of waterbodies to protect?

This is a tough question. The choice seems a bit arbitrary. It is key that those bodies that are naturally acidic and would not benefit from reductions in deposition have already been excluded. Protecting only half the sensitive water bodies seems clearly like not enough. What percentage of water bodies in the Adirondacks are currently affected? It is already established that current effects are adverse?

d) alternative standards for NOx and SOx…taking into account target ANC, target percentages of water bodies protected, relevant uncertainties, other factors such as co-protection?

The question of how to group resources seems an important one that needs to be resolved. At a minimum the split into two categories seems necessary. It is not clear that the benefits of going to the ecoregion level are worth the extra effort. A key question is whether further disaggregation would put less restriction on locations that are not sensitive—which is the whole reason why something other than a uniform national standard is being developed.

Comments from Dr. Ellis Cowling

Charge Question 24. In light of the Panel's views on what constitutes adverse effects to public welfare (see Chapter 3), what are the Panel's views on:

a) the degree to which current levels of NOy and SOx deposition are adverse to public welfare based on evidence and risk information, and information on adversity provided in Chapters 2, 3, and 4?

The ISA and REA for the current review of the NAAQS for oxides of nitrogen and sulfur (as summarized in Chapters 2 and 3) make very clear that current levels (ambient concentrations) of air-borne nitrogen and sulfur compounds (that include not only NOy and SOx, as asked about in this Charge Question, but also include NHx and some as yet poorly characterized organic forms of nitrogen -- which I would abbreviate RHx) – see page 7-35) are now causing significant "disruptions in the structure and function of aquatic ecosystems" in various acid-sensitive regions of the US.

In this connection please note especially the following paragraphs in Chapter 2 page 2-3:

> *"The scientific evidence is sufficient to infer a causal relationship between acidifying deposition and effects on biogeochemistry and biota in aquatic ecosystems* (ISA 4.2.2). The strongest evidence comes from studies of surface water chemistry in which acidic deposition is observed to alter sulfate and nitrate concentrations in surface waters, the sum of base cations, ANC, dissolved inorganic aluminum and pH. (ISA 3.2.3.2). Consistent and coherent documentation from multiple studies on various species from all major trophic levels of aquatic systems shows that geochemical alteration caused by acidification can result in the loss of acid sensitive biological species (ISA 3.2.3.3). For example, in the Adirondacks, of the 53 fish species recorded in Adirondack lakes about half (26 species) were absent from lakes with pH below 6.0 (Baker et al., 1990b). Biological effects are linked to changes in water chemistry including decreases in ANC and pH and increases in inorganic Al concentration."

Chapter 3 also makes clear that although the Clean Air Act provides a very broad definition of different kinds of air-pollution-induced "effects" on public welfare, the Act in fact does not define "public welfare" as such, and also does not define "adversity to public welfare." Nevertheless EPA has historically interpreted air-pollution-induced "adversity" to include "disruptions in ecosystem structure and function" that are regarded as important to the people of this country. This working definition of "adversity" seems very sensible to me.

Chapter 3 also includes a brief introduction to the concept of "Ecosystem Services" and describes various economic valuation and "Willingness to Pay" (WTP) studies that show very clearly that many citizens of our country are willing to pay the administrative and operational costs of both private-sector and public-sector efforts to decrease the presently ongoing acidification of freshwater lakes and streams in such places as the Adirondack Mountains of New York and New England and the Shenandoah National Park in the eastern US and in acid-sensitive landscapes such as the grasslands of Minnesota and Coastal Sage Scrub (CSS) areas of California.

Chapter 4 makes very clear that the current NAAQS standards for oxides of nitrogen and sulfur are not adequate to protect sensitive aquatic and terrestrial ecosystems from acidification- and nutrient-enrichment effects induced by atmospheric deposition of total reactive nitrogen and sulfur compounds. See especially Chapter 4 page 4-2:

> " … the current standards are not directed toward depositional effects, and none of the elements of the current NAAQS – indicator, form, averaging time, and level – are suited for addressing the effects of [total reactive] N and S deposition. Thus, by using atmospheric NO2 and SO2 concentrations as indicators, the current standards address only a fraction of total atmospheric NOX and SOX, and do not take into account the effects from deposition of total atmospheric NOX and SOX. By addressing short-term concentrations, the current SO2 standards, while protective against direct foliar effects from gaseous SOX, do not take into account the findings of effects in the ISA, which notes the relationship between annual deposition of S and acidification effects which are likely to be more severe and widespread than phytotoxic effects under current ambient conditions, and include effects from long term deposition as well as short term."

Thus my response to Charge Question 24a is:

> Based on the evidence and risk information as well as the information on adversity provided in Chapters 2, 3, and 4, and in light of my professional views about what constitutes adverse effects to public welfare, I conclude that current atmospheric deposition loads of total reactive nitrogen and sulfur (including NOy, SOx, NHx, and probably RHx as well) are causing very substantial and publicly unacceptable adverse effects on public welfare in various parts of the US.
>
> I also believe that the AAPI approach currently being developed through the currently proposed and well-integrated "two criteria-pollutant" approach (with acidifying NHx emissions and deposition also being taken "as given") is well grounded in the present state of scientific understanding about acidification effects on aquatic ecosystems.
>
> In addition I believe that the present focus on adverse effects in aquatic ecosystems will very likely provide some important co-benefits with regard to decreased adverse acidification effects and decreased nutrient-enrichment effects in sensitive terrestrial and estuarine ecosystems as well as decreased air concentrations of methyl mercury.

b) target values for ANC that protect against adversity to public welfare in light of the information presented in Chapter 5 concerning levels of ANC and the ecosystem effects associated with those target ANC levels?

Thus, my response to Charge Question 24b is:

> I regard the three suggested target values outlined in Chapter 5 for use of ANC as the ecological indicator of choice – 20 μeq/L, 50 μeq/L, and 100 μeq/L – to be very reasonable alternative levels for the Administrator of EPA to use in making her final decisions about the target value of ANC that would be appropriate for various acid-sensitive regions of our country.

c) factors relevant in selecting target percentages of water bodies to protect at alternative target ANC levels to protect against adverse effects to public welfare, and weights to place on those factors?

Thus, my response to Charge Question 24c is

> The addendum we received on September23 provided some clarification of "factors relevant in selecting target percentages of water bodies to protect at alternative ANC levels" and some information about "weights that could be placed on these factors" but after careful and repeated rereading of this addendum and other parts of Chapter 5, I still am not able to figure out how to formulate an appropriate response to Charge Question 24c, other than the obvious idea that protecting 90% of the water bodies would be more stringent than protecting 75% of the water bodies, and that protecting only 50% of the water bodies would be even less stringent.

d) alternative standards for NOx and SOx that would protect against adverse effects to public welfare based on the AAPI form, and taking into account
(i) consideration of target levels of ANC (chapter 5),

Chapter 5 describes the range of ANC values that are necessary to both understand and then make decisions about protection of freshwater lakes and streams from acidification caused by atmospheric deposition of total reactive nitrogen and sulfur:

- Water bodies with ANC values near or above 100 μeq/L have little or no risk of acidification,
- Water bodies with ANC values between 100 and 50 μeq/L are at progressively increasing risk of acidification,
- Water bodies with ANC values between 50 and 20 μeq/L are at even greater risk of acidification, and
- Water bodies with ANC values of 20 μeq/L or lower already are so acidic that most of them will not support viable populations of fish and many other aquatic biota.

Thus my response to Charge Question 24d(i) is:

> The NOx and SOx standards that will be necessary to protect water bodies in an acid-sensitive region of the US will be an inverse function of the ANC values already existing in the population of water bodies that are to be protected – i.e., the lower the existing ANC values in the water bodies to be protected, the more stringent must be the NOx and SOx standards that must be met.
>
> In addition, the final SOx standards that are established for each acid-sensitive region that is to be protected will need to be adjusted in part by determinations of the percentage of sensitive water bodies in the region that are to be protected and also by calculations or measurements of the nitrogen-assimilative capacity of the ecosystems that are to be protected.

(ii) target percentage of water bodies to protect (chapter 5),

My response to Charge Question 24d(ii) is essentially the same as my response to Charge Question 24b (above)

> The addendum we received on September23 provided some clarification of "factors relevant in selecting target percentages of water bodies to protect at alternative ANC levels" and some information about "weights that could be placed on these factors" but after careful and repeated rereading of this addendum and other parts of Chapter 5, I still not figure out how to formulate an appropriate response to Charge Question 24c, other than the obvious idea that protecting 90% of the water bodies would be more stringent than protecting 75% of the water bodies, and that protecting only 50% of the water bodies would be even less stringent.

(iii) consideration of relevant uncertainties in AAPI components (chapter 7).

Chapter 7 provides a very succinct and thorough introduction to many of the still existing uncertainties that are inherent in the AAPI approach to setting welfare-based NAAQS standards for NOx and SOx. As Chapter 7 and both the earlier ISA and REA documents make clear, however, major advances have been made in recent years in decreasing many of the scientific uncertainties that were considered in previous NAAQS reviews for NOx and SOx. Thus, a much more robust scientific foundation has been developed for establishing NOx and SOx NAAQS standards that will diminish the frequency and intensity of nitrogen and sulfur induced adverse effects on the structure and function of ecosystems and on ecosystem services important to public welfare in this country.

These important decreases in scientific uncertainty have resulted from the following developments in recent years:

1) The decision to take a two-criteria pollutant (nitrogen and sulfur) integrated approach rather than to continue to consider NOx and SOx separately,

2) Separating the development of public-welfare-based NAAQS standards from the formerly always dominating public-health-based NAAQS review processes,

3) Including in the AAPI approach to management of acidifying nitrogen and sulfur deposition, both chemically oxidized and chemically reduced inorganic forms of nitrogen (and even recognizing that organic as well as inorganic forms of nitrogen also must be considered) in the current ecosystem-focused secondary NAAQS review process,

4) Considering both acidification effects and nutrient-enrichment effects on whole ecosystems (including interactive effects among all types of plants, animals, insects, and microorganisms) rather than just direct effects on individual species of plants and/or animals,

5) Focusing on nitrogen and sulfur effects on naturally occurring and unmanaged terrestrial and aquatic ecosystems (that include natural grasslands; open range lands; unmanaged coniferous, hardwood, and mixed-species forests; and riverine, estuarine, and coastal ecosystems -- rather than trying also to consider at the same time, air-borne nitrogen and sulfur effects on commercially important plant and animal agricultural production systems and intensively managed commercial forests,

6) Greatly improved mathematical models (especially CMAQ) of spatial and temporal relationships among air emissions of pollutants, meteorological transport phenomena, chemical and physical transformations of airborne nitrogen and sulfur compounds, and wet, dry, and occult (cloud and fog) deposition processes at both high and low elevations,

7) Greatly improved concepts and descriptions of the diversity array of eco-regions that exist across this great continent of ours,

8) Much improved understanding of linkages among bed rock geology, soils, vegetative cover, temperature and moisture-supply gradients, episodic phenomena such as droughts, floods, snow melt processes, physical climate process, and chemical-climate-induced changes in the physical climate,

9) Recognition that our present scientific understanding of nitrogen- and sulfur-induced acidification and nutrient-enrichment processes in aquatic ecosystems is much more thoroughly developed than acidification and nutrient-enrichment phenomena in terrestrial and estuarine ecosystems.

Thus, my response to Charge Question 24d(iii) is:

> Yes, there are still some important uncertainties about how many different categories of sensitivity to aquatic ecosystems should be recognized, how adequately the estimates of chemically reduced forms of nitrogen from the CMAQ air quality model can be trusted, how large the co-benefits for terrestrial ecosystems will be from use of the present AAPI approach with its primary focus

on protection of aquatic ecosystems, and in the several kinds of Willingness to Pay (WTP) and other kinds of benefit estimates, but I am confident that these and other sources of uncertainty will continue to decrease during the next few years and that the present scientific foundation is adequate to implement the AAPI approach as soon as final decisions about the indicator, level, statistical form, and averaging time of the proposed NAAQS standard can be resolved.

(iv) any other potentially relevant factors, such as levels of co-protection against terrestrial acidification and nutrient enrichment (chapter 6)?

Chapter 6 contains a very short but persuasive description of the likelihood of significant co-benefits in protection of terrestrial ecosystems from acidification and nutrient enrichment effects from implementation of an AAPI approach aimed primarily at protection for aquatic ecosystems.

Thus, my response to Charge Question 24d(iv) is:

> Although it is difficult to develop quantitative estimates of the co-benefits that are likely to accrue in terrestrial ecosystems from NAAQS standards designed specifically to diminish adverse effects on aquatic ecosystems in acid-sensitive regions, I believe there is no uncertainty at all that such co-benefits will occur and will not be surprised if these co-benefits turn out to be significant in magnitude.

Chapter 4: Addressing the Adequacy of the Current Standards

Charge Question 2. What are the Panel's views on staff's approach to translating the available evidence and risk information and other relevant information into the basis for reaching conclusions on the adequacy of the current standards and on alternative standards for consideration?

My response to Charge Question 2 is essentially the same as my response to Question 24a:

> "Chapter 4 makes very clear that the current NAAQS standards for oxides of nitrogen and sulfur are not adequate to protect sensitive aquatic and terrestrial ecosystems against acidification and nutrient enrichment induced by atmospheric deposition of total reactive nitrogen and sulfur compounds.
>
> In this connection, please note especially Chapter 4 page 4-2:
>
> " … the current standards are not directed toward depositional effects, and none of the elements of the current NAAQS – indicator, form, averaging time, and level – are suited for addressing the effects of [total reactive] N and S deposition. Thus, by using atmospheric NO2 and SO2 concentrations as indicators, the current standards address only a fraction of total atmospheric NOX and SOX, and do not take into account the effects from deposition of total atmospheric NOX and

SOX. By addressing short-term concentrations, the current SO2 standard, while protective against direct foliar effects from gaseous SOX, does not take into account the findings of effects in the ISA, which notes the relationship between annual deposition of S and acidification effects which are likely to be more severe and widespread than phytotoxic effects under current ambient conditions, and include effects from long term deposition as well as short term."

a) In light of the Panel's views on the appropriate definitions of adversity to public welfare (see Chapter 3), do you agree that the current levels of NOy and SOx deposition are adverse to public welfare?

Yes, I do agree that the current levels of NOy and SOx deposition are adverse to public welfare. Once again let me explain my response by repeating parts of my response to Charge Question 24a:

Chapter 3 makes clear that although the Clean Air Act provides a very broad definition of different kinds of air-pollution-induced "effects" on public welfare, the Act in fact does not define "public welfare" as such, and also does not define "adversity to public welfare." Nevertheless EPA has historically interpreted air-pollution-induced "adversity" to include "disruptions in ecosystem structure and function" that are regarded as important to the people of this country. Thus EPA's working definition of "adversity" seems very sensible to me.

The ISA and REA for the current review of the NAAQS for oxides of nitrogen and sulfur (as summarized in Chapters 2 and 3) make very clear that current levels (ambient concentrations) of air-borne nitrogen and sulfur compounds (including not only NOy and SOx, as asked about in this Charge Question (but also include ambient NHx and some as yet poorly characterized organic forms of nitrogen (see Chapter 7 page 7-35) are now causing significant "disruptions in the structure and function of aquatic ecosystems" in various acid-sensitive regions of the US.

In this regard, please note especially Chapter 2 page 2-3:

"The scientific evidence is sufficient to infer a causal relationship between acidifying deposition and effects on biogeochemistry and biota in aquatic ecosystems (ISA 4.2.2). The strongest evidence comes from studies of surface water chemistry in which acidic deposition is observed to alter sulfate and nitrate concentrations in surface waters, the sum of base cations, ANC, dissolved inorganic aluminum and pH. (ISA 3.2.3.2). Consistent and coherent documentation from multiple studies on various species from all major trophic levels of aquatic systems shows that geochemical alteration caused by acidification can result in the loss of acid sensitive biological species (ISA 3.2.3.3). For example, in the Adirondacks, of the 53 fish species recorded in Adirondack lakes about half (26 species) were absent from lakes with pH below 6.0 (Baker et al., 1990b). Biological effects are linked to changes in water

chemistry including decreases in ANC and pH and increases in inorganic Al concentration."

3. Has staff appropriately applied this approach in reviewing the adequacy of the current standards and potential alternative standards?

Yes, I believe that EPA staff has very appropriately noted that:
1) the very short-term present secondary NOx and SOx standards (calculated as the arithmetic mean of 1-hour concentrations of NO_2 and as the arithmetic mean of 3-hour concentrations of SO_2) are wholly inadequate to protect aquatic or terrestrial ecosystems from the long-term cumulative acidifying loads of total reactive nitrogen and sulfur compounds;
2) the "indicators" used in the present NAAQS standards do not include all of the acidifying and nutrient-enriching forms total reactive nitrogen and sulfur that are now causing significant adverse impacts on the structure and function of aquatic and terrestrial ecosystems in various acid-sensitive regions of the US.

4. Has staff appropriately acknowledged the potential beneficial effects of nitrogen inputs into nutrient limited ecosystems, while maintaining the focus of the review on preventing adverse effects in nitrogen sensitive ecosystems?

Yes, in this connection please note the following discussion in Chapter 4 pages 4-44 and 4-45:
"In certain limited situations, additions of nitrogen can increase rates of growth, and these increases can have short term benefits in certain managed ecosystems. As noted earlier, this review of the standards is focused on unmanaged ecosystems. As a result, in assessing adequacy of the current standards, we are focusing on the adverse effects of nutrient enrichment in unmanaged ecosystems. However, the following discussion provides a brief assessment of effects in managed ecosystems.

Impacts of nutrient enrichment in managed ecosystems may be positive or negative depending on the levels of nutrients from other sources in those areas. Positive effects can occur when crops or commercial forests are not receiving enough nitrogen nutrients. Nutrients deposited on crops from atmospheric sources are often referred to as passive fertilization. Nitrogen is a fundamental nutrient for primary production in both managed and unmanaged ecosystems. Most productive agricultural systems require external sources of nitrogen in order to satisfy nutrient requirements. Nitrogen uptake by crops varies, but typical requirements for wheat and corn are approximately 150 kg/ha-yr and 300 kg/ha-yr, respectively (NAPAP, 1990). These rates compare to estimated rates of passive nitrogen fertilization in the range of 0 to 5.5 kg/ha-yr (NAPAP, 1991).

Chapter 6: Co-protection for Other Effects Using Standards to Protect Against Aquatic Acidification

Charge Question 13. What are the Panel's views on the utility of the additional analyses of co-protection benefits to inform consideration of alternative levels of the standard?

> My view is that the additional analyses of co-protection benefits contained in Chapter 6 is a well-reasoned and valuable addition to this Policy Assessment Document. I was especially well-pleased with the following summary paragraph in Chapter 6 page 6-2 and the additional detailed information contained in Tables 6-1 and 6-2 on page 6-3:
>
> "Results of the comparison between the aquatic critical acid load (ANC = 50 μeq/L) and
> the terrestrial critical acid loads (Bc:Al 1.2 and 10.0) for the 32 watersheds are presented in Tables 6.1 and 6.2. In the 16 Adirondack watersheds, 13 of the 29 lakes had aquatic critical acid loads that were lower (more protective) than the terrestrial critical acid loads when a Bc:Al ratio of 10.0 was used. Based on terrestrial critical acid loads determined with a Bc:Al ratio of 1.2, 21 of the 29 lakes in the Adirondacks had aquatic critical acid loads lower than the terrestrial critical acid loads. More importantly, for the terrestrial critical acid loads determined with a Bc:Al ratio of 10.0, 13 of the 16 lakes in the Adirondacks classified as "highly" and "moderately" sensitive to acidification had aquatic critical acid loads lower than the terrestrial critical acid loads, and all 16 lakes in these two sensitivity classes had critical acid loads lower than the terrestrial loads determined with a Bc:Al of 1.2 The watersheds within the Shenandoah region showed similar results (Table 6.1)."

Let me turn now to a few general remarks deriving from my experience as the designated "liaison person" serving as a member of both this NOx/SOx Secondary NAAQS Review Panel and the Integrated Nitrogen Committee (INC) developed within EPA's Science Advisory Board.

The first and perhaps most important linkage between the INC and the NOx/SOx Panel was the following "Resolution" developed by the INC and communicated to the NOx/SOx Panel on October 31, 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.

The NOx/SOx Panel has accepted this resolution and incorporated NOy as the recommended "indicator" of choice for implementation of the proposed revision of the NOx and SOx Public Welfare based NAAQS standards using the AAPI approach.

The second important linkage between the INC and the NOx/SOx Panel was a presentation in September 2008 by Chairman Russell of the then emerging AAPI approach, with its incorporation of chemically reduced forms as well as chemically oxidized forms of reactive nitrogen as an "as given" feature of regions of the US to which the AAPI approach could be applied. This novel approach was useful in developing an integrated way of recognizing that chemically reduced inorganic forms of nitrogen (gaseous NH_3 and ammonium ion (NHx^+) as well as chemically oxidized forms of reactive nitrogen and sulfur ($NOy+SO_2+ SO_4$) are all very important parts of the total acidifying deposition that leads to adverse ecosystem impacts in acid-sensitive regions of the US.

This choice to include NHx "as given" in the AAPI index for ecosystem effects of oxides of nitrogen and sulfur was is an artful means of avoiding the large administrative and probably nearly prohibitive political challenges of trying to designate ammonia and ammonium ion as a seventh Criteria Pollutant and thus including three rather than two Criteria Pollutants in this initial step that EPA is now taking in exploring options for multi-pollutant approaches in air quality management in the US as recommended in the National Research Council's 2004 report on "Air Quality Management in the United States."

The third important linkage between the NOx/SOx Panel and the INC came about during EPA's renegotiation of the original court-ordered deadline for completion of the NOx/SOx NAAQS review process. This change in the court-ordered deadline provided approximately 18 moths of additional time that EPA Staff sorely needed to complete the additional analyses and assessments that we presently have available in this Second External Review Draft Policy Assessment.

The fourth and last important linkage between scientific findings and recommendations from the INC and the findings and recommendation of the NOx/SOx Panel has to do with the magnitude of air emissions from various US sources of the reactive nitrogen and sulfur. As indicated in the attached Table 2 from the June 2010 draft report of the INC, in 2002 the total air emission of reactive nitrogen from industrial and transportation sources totaled about 6.2 Tg of NOx-N compared to agricultural sources that totaled about 3.1 Tg/yr of NHx-N – roughly a two-fold difference in air emissions of total reactive nitrogen from these three major sources.

Table 1: Nr fluxes for the United States, Tg N in 2002.[a]

Nr inputs to the *Atmospheric* environmental system			Tg N/yr	%
	N_2O-N emissions [1]		0.8	8
		Agriculture - livestock (manure) N_2O-N	0.03	

		Agriculture – soil management N_2O-N	0.5	
		Agriculture - field burning agricultural residues	0.001	
		Fossil fuel combustion - transportation*	0.1	
		Miscellaneous	0.1	
	NH_x-N emissions [2]		3.1	31
		Agriculture: livestock NH_3-N	1.6	
		Agriculture: fertilizer NH_3-N	0.9	
		Agriculture: other NH_3-N	0.1	
		Fossil fuel combustion – transportation *	0.2	
		Fossil fuel combustion - utility & industry *	0.03	
		Other combustion	0.2	
		Miscellaneous	0.1	
	NO_x-N emissions [2]		6.2	61
		Biogenic from soils	0.3	
		Fossil fuel combustion – transportation *	3.5	
		Fossil fuel combustion - utility & industry *	1.9	
		Other combustion	0.4	
		Miscellaneous	0.2	
	Total *Atmospheric* inputs		10.0	100
Nr inputs to the *Terrestrial* environmental system				
	Atmospheric N deposition[b]		6.9	19
		Organic N [3]	2.1	
		Inorganic NO_y-N [4]	2.7	
		Inorganic-NH_x-N [4]	2.1	
	*N fixation in cultivated croplands [5]		7.7	21
		Soybeans*	3.3	
		Alfalfa*	2.1	
		Other leguminous hay *	1.8	
		Pasture*	0.5	

		Dry beans, peas, lentils *	0.1	
	N fixation in non-cultivated vegetation * [6]		6.4	15
	N import in commodities *[7]		0.2	0.3
	Synthetic N *[8]		15.1	41
		Fertilizer use on farms & non-farms	10.9	
		Non-fertilizer uses	4.2	
	Manure N production [9]		6.0	16
	Human waste N [10]		1.3	3
	Total *Terrestrial* inputs		43.5	100
Nr inputs to the *Aquatic* environmental system				
	Surface water N flux [11]		4.8	

Also attached please find the following Concluding Statement from the June 2010 draft report from the INC:

Concluding Statement

Fossil fuel combustion and food production have significantly increased the introduction of Nr (reactive nitrogen) into the US environment and, while there have been 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 [educationally focused] program.

Comments from Dr. Charles Driscoll

Executive Summary

ES-3, paragraph 4	What is meant by balance of base cations? Change to "dissolved inorganic aluminum."
ES-3, paragraph 5, line 2	Change to "depth of soil and surficial deposits"
ES-4, paragraph 2, line 4	ANC of 50 μeq/L
ES-4, paragraph 4, line 3	Change to "may result in <u>nutrient</u> imbalance"
ES-4, paragraph 6	Change to "as trout are <u>eliminated</u> due to acidification"
ES-7, paragraph 2	Why would watersheds with low base-cation weathering be eliminated from consideration? These should be the most acid-sensitive watersheds.
ES-10-11, 1st paragraph ES-11	I don't believe this statement on naturally acidic ecosystems is true. There are surface waters that are naturally acidic due to low rates of base cation supply and/or high inputs of naturally occurring organic acids. However, these systems can also be impacted by elevated inputs of acidic deposition. This is a widespread occurrence in the Adirondack region of New York. These naturally acidic surface waters will exhibit loss of ANC and elevated aluminum concentrations from acidic deposition.
ES-13, last bullet	This statement is problematic. Brook trout is not a sensitive species. Maybe the sentence should be changed to state "…protection against declines in fitness of <u>less</u> sensitive species (e.g., brook trout, zooplankton) …" Also, what is meant by "the overall health of aquatic communities may not be impacted." If species are lost, isn't this an impact on the health of aquatic communities? I think this bullet needs to be re-phrased.

Chapter 1

P 1-9, line 4-6	Is this sentence correct? Aren't both direct and indirect effects considered in this PA? Also, total deposition is not just particulate forms. This sentence should be re-written.
Chapter 2	
P 2-2, line 8	This sentence needs to be changed to something like "in some instances unless strongly retained by soil or biota, leach out …"
P 2-9 in PnET box	Change last line to "The model can be set to operate on any time set, but is generally run on a monthly time-step. It is applied at the stand to small-watershed scale."
P 2-21, line 1	It might be good to reference Goodale et al. (2010) here.
P 2-21, paragraph 2	The article by Thomas et al. (2010) on nitrogen deposition on northern tree species should be mentioned in this paragraph.
Chapter 3	
P 3-6, line 13-15	This statement about alkalinity is incorrect. For all intents and purposes, alkalinity and ANC are the same. Often alkalinity involves titration to a fixed pH endpoint (~4.2), while ANC generally involves Gran Plot determination of the equivalence point. The difference between the two is subtle at best. For this document, the two should be used interchangeably.
P 3-9, line 11	This sentence on the units conversion does not make sense. Sulfur and nitrogen have different molecular weights. Therefore one cannot have a single mass conversion for a nitrogen map (left) and a sulfur map (right).
P 3-15, Figures 3-5 and 3-6	Does the N deposition include NH_4^+, is the map total N deposition or NO_3 deposition? Please clarify.
Page 3-30, line 8	100 µeq/L

Chapter 4

P 4-1, line 6	Change to "associated with elevated deposition of NO_x …"
P 4-1, line 22	Change to "nutrients and acid neutralizing capacity …"
P 4-2, line 25	Change to "as the ability of the watershed to counteract acidic inputs is decreased as the supply of acid neutralizing capacity is used more rapidly than can be replaced through geological weathering."
P 4-20, line 2	The text refers to sulfur fields but the figures referenced (Figure 4-5, 4-6) depict NO_x and NH_x. Is there a mistake here?
P 4-20, line 4	The text refers to concentration patterns, but Figure 4-4 shows deposition.
P 4-20, lines 24, 29	The text refers to correlation between NO_x and N deposition. Is this NO_x concentrations? If so, this should be clarified.
P 4-49, line 27	Change to "and methyl mercury can be taken up…"
P 4-50, line 14-15	Methylation of mercury occurs in watersheds all over the U.S. (and the world) where conditions are appropriate. Please change this sentence, it is incorrect.
P 4-50	Note there are other linkages between acidification and fish mercury accumulation. Mercury is accumulated to a greater degree in aquatic biota as pH and ANC decreases. Dittman and Driscoll (2009) noted that as fish condition increased associated with decreases in acidic deposition, fish mercury concentrations decreased.

Chapter 5

P 5-8, line 21-22	Change to "some fraction of the acid neutralizing capacity…"

P 5-13 (Figure 5-4b)	I am not familiar with this paper, but the figure does not make sense, why would the ANC curve have different lines for a wet US average year? This figure should be explained or deleted.
P 5-15, line 6	Change to “to neutralize the deposition.”
P 5-15, line 16	This definite of steady-state models is horrible. A steady state model is one with time invariant inputs, outputs and pools. This section should be re-written.
P 5-17, entire page	The authors are using the term equilibrium incorrectly. Equilibrium is a thermodynamic term. Throughout this page, the word equilibrium needs to be replaced with the word “steady-state” (lines 1, 4, 6, 8, 14, 29, 30).
P 5-17, line 16	There is a problem here with the term critical load. Critical load is a steady-state phenomenon. For a value of critical load that is not at steady-state, the term dynamic critical load or target load should be used.
P 5-17, line 24	Change to “implying that watersheds with greater inherent supply of acid neutralizing capacity respond …”
P 5-19, line 7	Change to “in-lake retention of SO_4^{2-} and N.
P 5-19 (around here)	A critical issue needs to be addressed if steady-state models are going to be used. Steady-state models will give relative high values of the level of atmosphere deposition needed to protect ecosystem (critical loads) because they assume steady-state conditions. At any reasonable time frame, the dynamic critical load will be much lower. If steady-state models are going to be used a “safety factor” should be applied to account for this discrepancy.
P 5-19, Figure 5-5	Does the trade-off figure consider background deposition? In other words, is the zero deposition value really 0? There is background deposition of S and N that will not be changed by controls of emissions.

P 5-21, equation 3	This equation does not make sense and needs to be explained better. What is the difference between nitrogen uptake and nitrogen immobilization?
P 5-23, lines 10, 16	Change to "as the acid neutralizing capacity of the watersheds increase…"
P 5-26, 5-27, line 18-19, Figure 5-7	The text refers to a map of critical loads, but Figure 5-7 is a map of sites where critical loads have been calculated.
P 5-27	This approach of eliminating low ANC sites seems foolish. These are the most sensitive sites. Why would you throw them out? If you don't want to include these sites, the percentage of the lakes targeted should be relaxed. This would be a more honest, transparent approach rather than throwing out the most sensitive watersheds. Also I would recommend against throwing out the high DOC lakes. High DOC waters can be impacted by acidic deposition. A better approach would be to include these waters and check the DOC concentrations of the waters that would not be protected. Undoubtedly these would include many high DOC waters.
P 5-51, Figure 5-18	I would like some additional explanation of this figure. It appears that the NH_x deposition shifts the "dog-leg" to lower values of N deposition for graphs a and c, but not b. Why?
P 5-52, Figure 5-19	This figure is also difficult to follow. I think I have the sense of it, but a more detailed explanation would be helpful.
P 5-54, line 14	What is meant by pure nitrogen and sulfur? Do you mean total nitrogen and total sulfur? Please clarify.
P 5-54, line 26	Would better wording be "N and S atoms of NO_x and SO_x removed from the atmosphere, which …"
P 5-56, line 8	Would better wording be "species that affect the health of ecosystems would…"
P 5-56, line 25	Should you specify which lake in the Adirondacks?

P 5-58, Figure 5-20	This figure suggests that T values are relatively invariant for the eastern U.S. Is this correct? If so, this would be important information to clarify. It would also be helpful to explain why this is the case (also discussed in Chapter 7).
P 5-65, line 6	It is important to define what is meant by uptake and immobilization. It also would be helpful to indicate how uptake, denitrification and immobilization are calculated.
P 5-66, line 23	Rather than nitrogen buffering capacity, do you mean nitrogen retention capacity?
P 5-66, line 24	Again this is confusing. Do you mean "when reduced nitrogen deposition exceeds the ability of the ecosystem to retain nitrogen?
P 5-66, 2nd paragraph	This paragraph is confusing and needs to be re-worded. The term buffering capacity is not being used properly. There is confusion on distinguishing between nitrogen retention and loss of acid neutralizing capacity.
P 5-67, line 3	Clarify units 50 μeq/L.
P 5-70, paragraph ????	This paragraph is horrible. For example, line 7 indicates that below pH 4.5 ANC appears to be uncorrelated with pH. As at pH values below the equivalence point ANC = - $[H^+]$ this shows what an incorrect statement this is. This paragraph is filled with mis-statements and needs to be completely re-written.
P 5-75, Table 5-10	Aren't these species listed from most sensitive to least sensitive? See table title.
P 5-81, line 13	How about "as ANC decreases, the probability of very low pH values occurring increases."
P 5-82, line 24	Brook trout is a relatively insensitive fish species.
P 5-82, line 25	How about "When ANC values are <50 μeq/L, the probability of acidic episodes increases substantially."

P 5-82, line 30	How about "At these levels during acidic episodes brook trout populations…"

Chapter 5 General Comments (the devil is in the details)

- How will the probability of lakes to be protected be determined?
- How many sites per region/category will be evaluated?
- How will the time-dependence of recovery be addressed? Critical loads vs. target loads (dynamic critical loads).

Chapter 6

P 6-1	This analysis is nice but I am skeptical. There are limited field observations on this. Many soil time series studies over the past 15 years show ongoing depletion of soil exchangeable calcium and magnesium which many waters, particularly in the Northeast, are showing recovery of ANC. This pattern, if true, suggests ongoing soil acidification while surface waters are recovering from acidic deposition. This may also suggest that soil is more "sensitive" to inputs of acidic deposition than surface waters.
P 6-4, line 17	50 μeq/L
P 6-6	How about a short blurb about co-benefits associated with decreases in fish mercury and wildlife mercury concentrations associated with decreases in sulfate loading and/or increases in surface water pH?

Chapter 7

P 7-13, 7.4.3.2	It would be helpful and important to discuss why T values are relatively homogenous.
P 7-17-7-20, Figures 7-1, 7-4	Indicate what the lines on the figures represent.
P 7-32, line 1	Change to "as the supply of acid neutralizing capacity of watersheds increases…"

References:

Dittman, J.A., Driscoll, C.T., 2009. Factors influencing changes in mercury concentrations in yellow perch (*Perca flavescens*) in Adirondack lakes. Biogeochemistry 93 (3), 179-196.

Goodale, C.L., Thomas, R.Q., Dentener, F., Adams, M.B., Baron, J.S., Emmett, B.A., Evans, C.D., Fernandez, I.J., Gundersen, P., Hagedorn, F., Lovett, G.M., Kulmatiski, A., McNulty, S.G., Melvin, A.M., Moldan, F., Ollinger, S.V., Schleppi, P., Weiss, M.S., In press. Nitrogen deposition and forest carbon sequestration: A quantitative synthesis from plot to global scales. Global Change Biology.

Thomas, R.Q., Canham, C.D., Weathers, K.C., Goodale, C.L., 2010. Increased tree carbon storage in response to nitrogen deposition in the US. Nature Geosciences 3, 13-17.

Comments from Dr. Christopher Frey

Chapter 7: General Comments

Chapter 7 should have an introduction regarding the spatial scope and temporal averaging that is the basis for each of the inputs or factors discussed in the chapter. Without a context regarding spatial scope and temporal averaging, it is not possible to characterize either variability or uncertainty. It needs to be clear, for example, as to over what geographic domain and for what averaging time quantities such as deposition transformation ratios are to be estimated.

Charge Question 16: What are the Panel's views on the discussion of uncertainty in the critical loads models including MAGIC and SSWC?

The chapter discusses the MAGIC critical load simulations on pages 7-3 to 7-4, page 7-6, and in three paragraphs on pages 7-30 to 7-31. The discussion is with respect to an uncertainty analysis conducted in the REA. Figures 7-14 and 7-15 illustrate that the MAGIC model estimates for surface water chemistry at two locations compared very well with observed values. The simulated values fall very close to the parity line, demonstrating that the model is both precise and accurate in estimating concentrations, ANC, and pH.

On lines 8-9 on page 7-31, the text is "The estimated confidence bounds on predicted ANC suggest that the 95 percent upper confidence bound is on average 10 percent higher in lakes, and 5 percent higher in streams." It is not clear as to what is being compared – higher than what? Is the intended meaning that the 95 percent upper confidence bound is 10 percent higher than the mean value? Or 10 percent higher than the observed value?

For MAGIC, given that there are comparisons of model predictions to observed values that demonstrate the model performance, the discussion appears to be adequate.

The discussion of uncertainty in the SSWC makes several points:

- The F-factor approach is widely used in Europe and Canada, but not in the U.S.
- Critical loads estimated by steady-state MAGIC and SSWC F-factor approaches had similar trends and results converged for low critical loads
- In the REA, a Monte Carlo-based uncertainty analysis was conducted by varying runoff rates, water chemistry variables, and acid deposition. The coefficients of variations were 5 to 9 percent for critical load limits of 20 to 50 ueq/L.

The statement that the uncertainties introduced by the SSWC F-Factor model are likely to be moderate seems to be supported by the discussion in the text.

Charge Question 17: What are the Panel's views on the areas for future research and data collection outlined in this chapter, on relative priorities for research in these areas, and on any other areas that ought to be identified?

The chapter does not clearly outline areas for future research or data collection, nor does it offer priorities for research.

Perhaps this charge question refers to the section on modeling and data gaps. For consistency with the PA's for other criteria pollutants, consistent header titles should be used. If the purpose of this section is to identify areas for future research and data collection, there should be an introductory paragraph that states this.

From the current draft, one infers that the key needs for better models and data are:

- Occult deposition from cloud and fog processes, especially in high elevation watersheds
- Lightning generated NOx, to be incorporated into CMAQ in 2012
- The role of organic bound nitrogen in wet deposition, and re-entrainment from the surface
- Increased spatial coverage of ambient measurements, and specific measurement "needs" (better instruments? Could be more clear) related to NOy, speciated NOy, ammonia, and ammonium.
- Better emissions estimates for soil and agricultural emissions of [NOy? Or just ammonia and NOx?].
- More data for sensitive ecosystem areas, to have more even coverage geographically.

There is no discussion, based on Table 7-1, of the key sources of uncertainty other than gaps that should be areas for future research or data collection. Examples include:

- Pre-industrial base cation concentration is listed as having a high magniture of uncertainty and a high knowledge base uncertainty. Should this be a priority for future research or data collection?
- Dry deposition (generically – N and S species) is listed as having a medium magnitude of uncertainty and a medium-high knowledge base uncertainty.
- Ecological indicator to changes in the value of ecosystem services is listed as having "medium-high" magnitude of uncertainty, and a "negative" bias. Is there a need for better information here?

Not mentioned is what research is needed in order to improve the knowledge base for ecosystem effects other than aquatic acidification, such as terrestrial acidification and aquatic system nutrient enrichment.

In the recent CASAC PM Panel review of the PM policy assessment second draft, a fairly substantial list of research needs was identified with a recommendation that EPA hold a follow-up workshop to further refine the research agenda and consider how to implement research in order to improve the state of knowledge before the next review cycle.

Charge Question 21. What are the Panel's views on the overall characterization of uncertainty as it relates to the determination of an ecologically-relevant multi-pollutant standard for NOx and SOx?

This charge question is rather broad and it is not clear as to the specific focus. Chapter 7 appears to do a thorough job of listing many sources of uncertainty and providing qualitative characterization of each.

Charge Question 22. What are the Panel's views on the following:

a. The insights that can be gained into potential alternative additional secondary standards (using the AAPI form) by considering:

i. Information from studies on the relationship between mortality in aquatic organisms and pH and ANC?

ii. Information from studies on the relationship between fish health and/or biodiversity metrics and pH and ANC?

iii. Information on the relationship between pH, Al, and ANC?

iv. Information on target ANC levels identified by states and regions, as well as other nations?

Chapter 9 could more clearly, and very briefly, summarize what are the key hazards associated with aquatic acidification, and list or briefly describe the key adverse effects, while stating conclusions regarding the relationship between mortality in aquatic organisms, pH, and ANC. The chapter presumes that the reader is already familiar with the endpoint effects, and tends to focus only on the indicator.

Likewise, Chapter 9 could more clearly summarize the relationship between fish health and biodiversity metrics with respect to pH and ANC. This should be done qualitatively, for a lay reader, leaving the technical details to the earlier chapters.

Although Chapter 9 implies that the use of the indicator for acidic deposition to aquatic systems may afford some protection with respect to acidic deposition to terrestrial systems, there could be a paragraph that more clearly but succinctly addresses the significance of, and linkages between, pH, Al, and ANC.

There is no discussion in chapter 9 that I could find regarding comparison of target ANC levels among states, regions, or nations. Perhaps a paragraph could be provided that summarizes this, leaving the details to previous chapters.

b. The appropriate role of qualitative and quantitative characterizations of uncertainty in developing standards using the AAPI form?

EPA has appropriately taken a weight of evidence approach to hazard identification. EPA has done a reasaonable job in identifying and characterizing individual sources of uncertainty, as summarized in Table 7-1. Other committee members may have specific comments on those sources of uncertainty, or others that might be included. The discussion of model and data gaps in Chapter 7 was useful, but needs to be expanded (see earlier charge question).

A key conclusion from Chapter 7 may be worth repeating in Chapter 9 (page 7-37, lines 19-29):

"there is no apparent directional bias in the uncertainty regarding the biological, chemical and physical processes incorporated in the AAPI. From the perspective of valuation of ecosystem services, the estimates generally are believed to be biased low, meaning the values of reaching a target level of protection are underestimated. However, quantification of these values is perhaps the most uncertain of all aspects considered. Consequently, the level of the AAPI should be relatively high in a buffering context to account for the existence of uncertainties in several components. In addition to, but related to these uncertainties discussions, are considerations of time lag to reach a target level ANC due to ecosystem response dynamics, as well the uncertainties in the severity and prevalence of episodic events. Both of these considerations suggest support for an AAPI that is somewhat higher than the target ANC supported by the specific evidence and risk information."

It is not clear, for example, that the conclusion that the AAPI should be somewhat higher than the target ANC (nor is this statement itself particularly clear, because it implies that AAPI and ANC are the same quantity) has been taken into account in the discussion of Target ANC limits in Chapter 9.

c. The role of considerations regarding the relationship of the standard to:

i. the time trajectory of response, e.g. when specific ANC levels are likely to be realized given a specific level of the AAPI?

ii. the likelihood of damages to aquatic ecosystems due to episodic acidification events given a specific target for chronic ANC?

iii. the levels of co-protection for terrestrial ecosystems against acidification effects and the for aquatic and terrestrial ecosystems against effects of excess nutrient enrichment?

These questions go beyond my expertise, but they imply a question regarding short-term versus long term responses of ecosystems, and the dynamics of eco-system response, that do not appear to be discussed in Chapter 9. For example, if a 3 to 5 year averaging time is used, should the level be set so as to also be protective against short term (e.g., seasonal, or shorter episode) events? A related question is whether the modeling tools, such as steady-state MAGIC simulations, adequately account for the adverse effects associate with dynamic responses. As another reviewer suggests

(Dr. Driscoll), perhaps a safety factor is needed to account for differences in dynamic versus steady-state response.

There is discussion of the implications of the aquatic system acidification-based standard with regard to its effect on terrestrial acidification and nutrient enrichment, which seems adequate but domain experts may have more comment on this issue than do I.

General comments on Chapter 9

The summary of suggested options given in table form is very useful and should be incorporated into chapter 9.

The choice of averaging times and whether only such averages or used, or whether shorter term episodes will be considered, needs to inform the variability and uncertainty analyses of Chapter 7. This is because the range of variability and uncertainty depend on averaging time. Similarly, the geographic scope needs to be taken into account in the analysis of variability and uncertainty. The spatial options for components of the AAPI equation need to be further discussed or refer the reader to specific text earlier in the document.

This standard may be challenging to communicate to the public and the regulated community. Finding a way to state the basic ideas in qualitative language that a lay reader can understand would be very helpful.

Although Chapter 9 alludes to attributes of a standard, it seems to require referring back to previous chapters in order to have a complete picture. In communicating this to the Administrator and other stakeholders, the EPA staff have a significant challenge of how to present the indicator, averaging time, form, and level in a manner that is relatively easy to explain and complete without overwhelming the audience with details.

- For example, the summary table is helpful in defining the "indicator" but seems incomplete. Isn't AAPI actually being used as an indicator? Not just NOy and SOx?
- The rationale for the 3 year, 5, year, and other averaging times should be explained.
- The form of the standard seems to be expressed in a complex manner. The cogent information is that NOy and SOx are related through AAPI. This could be stated more succinctly in the summary table. Explanations of other points can be in footnotes or the text.
- While the AAPI equation is important, perhaps it can be a footnote if it is not actually considered to be an indicator.
- The spatial options should be illustrated graphically in Chapter 9. This chapter should be thought of as the synthesis material that any reader will go to for the bottom line of this policy assessment. Thus, it should be complete in terms of information needed to completely define a standard.

- If the spatial categories are only regarding non-air quality inputs to the AAPI equation, then perhaps this material is not critical to Chapter 9 as long as it is clearly laid out earlier in the policy assessment. Or, it can be in a different table. On the other hand, there is a need for clarity on the geographic scale and averaging time for each of the AAPI inputs. Possibly this material needs to be moved to Chapter 7.
- To be consistent with how NAAQS are structured, the summary table should include a section on "Level."

Comments from Dr. Paul Hanson

General Comments:

I approve of the suggested approach taken by EPA staff to retain the existing standards for phytotoxic direct effects of NO_x and SO_x, and the development of an additional standard based on the deposition of combined forms of N and S. The focus on aquatic acidification effects for that additional standard is appropriate to the available data. My expertise lies in the area of terrestrial ecosystems and I will not comment on the specifics of the proposed aquatic-acidification-based standard.

In most instances, the document does an appropriate job of clarifying that demonstrated effects associated with acidification and nutrient enrichment from N and S deposition are limited to 'sensitive' ecosystems. Nevertheless, in a number of places the document slips back into language that could be interpreted as inferring that all ecosystems will benefit from such a new standard. Such language should be corrected to ensure that the reader understands that protection afforded by the proposed new standards would be limited to sensitive ecosystems located in specific regions of the United States.

The executive summary and conclusion (Chapter 9) fail to provide a clear picture about the extent of total US lands subject to the benefits of the proposed standard.

Comments on selected Charge Questions:

What are the Panel's views on the definitions of adversity that are appropriate to consider in determining what constitutes adversity to public welfare relative to the NOx and SOx secondary standards?

Without a defined understanding of how ecosystem changes in species composition would proceed through natural succession processes in the absence of anthropogenic N and S inputs, it is difficult to understand when changes in species or biodiversity should be considered adverse. The text tends to suggest that any change from the status quo should be considered adverse. I am not in agreement with such a conclusion. Unfortunately, I am also not providing a definition of the boundary of acceptable vs. unacceptable rates of change. The document should deal with this issue. If species changes in lichens or grassland composition are to be proposed as metrics of adverse effects, an attempt needs to be made to characterize levels of change to be expected in non-polluted ecosystems.

Comments on specific sections of the document:

Executive Summary:

1. Line numbers should be added to this section to facilitate comments.
2. On Page ES-3 Paragraph 3 the word "often" might be removed. It is insufficiently quantitative to be informative. Its inclusion also tends to lead the reader to conclude that deposition of NO_X and SO_x routinely leads to leaching from watersheds, which then result in the acidification of aquatic systems. This may be the case in some, but not all watersheds throughout the US where deposition of N and S forms are largely retained.

3. Page ES-4 paragraph 3: The claim at the end of this paragraph should be referenced to the ISA or REA.
4. Page ES-5 fourth paragraph: I don't agree with the first sentence of this paragraph, but rather think most N-limited ecosystems would have the capacity to absorb or buffer incoming N and S forms to limit or avoid perturbation. The examples of species change driven by deposition are provided (I guess) as definitive examples of an adverse result without adequate justification of the rates of species change that could or should be considered adverse.
5. ES-7 last paragraph: Deposition velocity is not the rate of pollutant deposition. Deposition velocity times the concentration gradient is the rate of deposition.

Comments from Dr. Rudolph Husar

Chapter 5: Conceptual Design of the Standard

5.3.1 Conceptual Design of the form.

I like the revised Figure 5.2. Still, instead of bidirectional arrows, two rows of arrows, one heading left the other to the right would more clearly communicate the intent.

9. What are the Panel's views on the revised characterization of the deposition transference ratios (TNOyand TSOx)?

In principle, I do agree with the approach to derive the ambient concentration – deposition relationship for individual S and N species using CMAQ. The aggregation approach also makes sense.

A repeat of my past comment on nomenclature: The Deposition/Concentration IS an effective total wet+dry deposition velocity. So why the addition of the confusing 'Transference" term? The concepts and formulae are already messy enough, so why not keep it simple: Effective Total Deposition Velocity. On p 7-3 its 'transformation ratio' . On p 7-12 its "Ambient Concentration to Deposition Transformation Ratio"….etc.

Figure 5.23 is the same as Fig 5.4

10. What are the Panel's views on staff's conclusion that an averaging time of 3 to 5 years is appropriate given the AAPI form of the standard?

Agree that 3-5 years is adequate for the characterization of inter-annual variability of key measures and indicators.

However, I am not clear as to why and how the episodic events and damage (and the springtime 'acid shock') are dismissed as is done in Section 5.2. Where is the evidence that an annual average standard will be more protective than a companion short-term standard?

Chapter 7 Uncertainty

14. What are the Panel's views on the following?

a. The degree to which the chapter appropriately characterizes the potential role of information on uncertainty, sensitivity, and variability in informing the standards?

This version of the Policy Assessment came a long-long way in characterizing the uncertainty, sensitivity and variability. My main concern remains to be known systematic errors in CMAQ simulations and their inadequate inclusion in the uncertainty analysis.

b. The appropriateness and completeness of the evaluation of CMAQ model performance and sensitivity to critical inputs?

The overestimation of the SO2 is a significant error that is not being 'corrected for'. Is that correct? High SO2 will probably result in high (unverifiable) dry deposition. So, focusing and showing the propagation of this known error would make the uncertainty analysis more convincing. For instance a figure like Fig 7-5 for CMAQ-CASTNET comparison for SO2 could be very revealing.

c. The utility of the analyses of temporal and spatial variability in the deposition transference ratios (TNOyand TSOx)?

The spatio-temporal variability of the Effective Total Deposition Velocity makes sense, since it indeed varies considerably in space and time. However, the CMAQ calculations of the Dep/Conc ratio for S and N compounds in Fig 5-20 do not appear to have the right spatial texture. Near sources where dry deposition of gases dominate, Dep/Conc(dep velocity) should be significantly higher than in the far field where wet deposition dominates. I don't see that texture. Inverting T in the plots FIG. 5.2 makes the interpretation even more difficult.

15. What are the Panel's views on the insights provided by the AAPI sensitivity analysis including: a. The evaluation of elasticities of response? b. The multivariable ANOVA analysis?

The value of the sensitivity/elasticity analysis heavily depends on what magnitude of perturbations one assumes. Are the assumptions reasonable?? I don't know. The CMAQ SO2 concentration bias is a big, known systematic error. Is it properly incorporated in the uncertainty.

Comments from Dr. Dale Johnson

Charge Question 2: What are the Panel's views on staff's approach to translating the available evidence and risk information and other relevant information into the basis for reaching conclusions on the adequacy of the current standards and on alternative standards for consideration?

Response: The Second Review Draft focuses almost entirely on aquatic effects with the rationale that such effects are better known and better documented than terrestrial effects. This is certainly true. But this raises a question in my mind: is it the purpose of this document to provide evidence to support changing standards, more or less in a lawyerly fashion? Should this be the purpose? Or should the purpose of the document be to examine all the potential pluses, minuses, and potential unintended consequences of changing standards? In short, is this a mission? If so, is the review panel expected to sign on to the mission? These questions came to mind as I considered my response to Charge Question 4: many of the points that I have raised in the past as potential benefits of increased N deposition to forest ecosystems and C sequestration are now moot with the change in focus toward aquatic effects. I reiterate that I am in no way against changing standards to protect aquatic ecosystems, I am only trying to see that the approach to it includes a balanced assessment of the effects of such changes. If EPA does not do it, I am quite sure that someone else will.

Charge Question 2a) In light of the Panel's views on the appropriate definitions of adversity to public welfare (see Chapter 3), do you agree that the current levels of NOy and SOx deposition are adverse to public welfare?

Response: This almost becomes a philosophical issue. It is hard to conceive of an effect of some perturbation that does not have some adverse as well as some beneficial effect to public welfare, with the probable exception of Hg deposition. The example that comes to mind is agricultural fertilization, which is adverse to public welfare when it is done in excess and leads to groundwater nitrate pollution, yet on the other hand, it is certainly adverse to public welfare to preclude fertilization with the resultant substantial decline in crop and food production! There is little doubt that current levels of NOy (combined with NH4) and SOx deposition are having adverse effects on some sensitive ecosystems; how many of such ecosystems can be protected at what cost, and what are the magnitudes and importance of unintended consequences (such as forest ecosystem C balance or crop S fertilization) that might result from such protection, and how does this compare to the benefits of protecting these sensitive ecosystems? The question becomes one of assessing the balance between these two effects, and, while recognizing the considerable uncertainties in some unintended consequences, I do feel that further discussions along such lines will add considerable credibility to this document.

Charge Question 4: Has staff appropriately acknowledged the potential beneficial effects of nitrogen inputs into nutrient limited ecosystems, while maintaining the focus of the review on preventing adverse effects in nitrogen sensitive ecosystems?

Response: The staff acknowledged at various places in the document that some benefits of N deposition might occur in very limited circumstances in commercial forests. They do not mention the C balance issue, which could occur in any forest, although the inclusion of Climate in Table 3-1 implies this, and is certainly of more practical relevance than" Climate Control" or "Regulating Climate" as is now shown in the table. While dismissing the potentially positive effects of N fertilization in non-commercial forests, the staff does, however, spend a considerable amount of time considering exactly the same phenomenon in non-commercial forests of the southwestern US where increased growth probably provides unwanted fuel for the next wildfire. I find this to be unbalanced. In general, however, any benefits of N (or S) deposition to terrestrial ecosystems is far less relevant in this document than in previous ones in that the staff has limited their scope largely to aquatic effects, none of which (to my knowledge) are beneficial. The wording in the Executive Summary regarding the ISA is correct (p. 1-9, lines 7-9) in that "The ISA highlights the ecological effects ….. to sensitive ecosystems other than commercially managed forests and agricultural lands…" Thus, the consideration of any potential benefits to ecosystems which by definition are "sensitive" (implying sensitive to negative effects) becomes moot, irrelevant, and dismissable. However, I would note that the first part of the quote from the Clean Air Act in page 2-1, lines 7-9, does not necessarily dismiss any benefits, but simply addresses effects. It does indeed mention damage to property, etc in the middle part of the quote, but the beginning and end do not specify that only negative effects be considered (although this may well have been what was intended). In a nutshell, if the scope of this effort is now limited to aquatic effects, then the issue of potential benefits becomes nearly irrelevant. It is not irrelevant, however, in the larger scheme of things where C balance effects of N deposition are now being hotly debated in the literature and sure to come up at some later time if new standards are proposed.

Charge Question 11: What are the Panel's views on the preliminary staff conclusions regarding alternative target ANC levels that are appropriate for consideration and the rationale upon which those conclusions are based?

The ANC levels at which negative effects on aquatic systems occur appear to be well established and I see no problem with considering alternate target ANC levels in this context. The premise of the section on alternate levels of ANC does not directly assess whether a given AAPI standard will or will not achieve a certain target ANC because of uncertainties, but presumes that the target ANC levels are reached and discusses them in that context (p. 5-69, lines 9-16). I confess to some degree of confusion as to the logic in some of this section, for example, the discussion of alternative target ANC's and timing on p. 5-85, lines 2-8. Clarification of this logic would help this reader. I would also point out that the capacity (change in soil) and intensity (change in soil solution) considerations raised before should enter into this discussion, as changes in the intensity factor could be very rapid in response to changes in deposition whereas changes in the capacity factor could be as slow as envisioned here, if indeed they occur at all (for example, it seems very unlikely that soil exchangeable acidity will decline and base saturation will increase in response to a decrease in acid deposition inputs – soils in humid environments would not likely become more basic with time, but only continue to acidify at a slower rate). I also confess to some confusion as to the derivation of the DL factors discussed on page 5-88 and 5-89 – if they were explicitly defined by other than the general terms used here, I missed it, and these terms are not in the list of abbreviations and acronyms up front.

Charge Question 11 a) In light of the Panel's views on appropriate definitions of adversity to public welfare (see Chapter 3), what are the Panel's views on the appropriateness of the information related to adversity considered by staff in evaluating alternative target ANC levels?

Here I have nothing further to add in addition to what was stated above.

Charge Question 12: What are the Panel's views on the approaches considered by staff for assessing alternative target percentages of water bodies for protection at alternative ANC levels?

Again, it is not clear how this was done – the DL factors, which clearly are numerical indices of some kind, are not defined in pages 5-88 and 5-89, and thus I am unable adequately address this question. If they were defined (preferably in the form of equations) elsewhere, that should at least be clearly referenced here, and preferably formally defined again. It is unclear to me how the $DL_{\%eco}$ terms in Tables 5-12 and 5-13 were derived, and yet it seems to be a critical element of the assessment. This needs to be more clearly described.

Other Specific Comments

Table 1-1: It would probably be better to use SI units here, as nearly all journals demand these days.

p. 1-9, lines 7-8: I do not understand why ag systems and commercial forest systems should be left out.

p. 2-1, line 24: need a space between "9" and "of"

p. 2-2, lines 13-28: Again, the intensity effects need to be included here – that is, introduction of strong acid anions such as sulfate and nitrate to an already acid soil (and acid soils do occur in nature, without any air pollution effects), then acidification of waters can occur instantly without any change at all in base saturation.

p. 2-5: Some discussion of natural acidification processes by natural carbonic and organic acids and by plant uptake should be discussed here. The uninitiated may erroneously conclude that acid soils only occur in the presence of air pollution.

p. 2-19, lines 20-21: This statement seems to fly in the face of other statements later on which say that there it too much uncertainty in terrestrial effects and therefore the document will concentrate on aquatic effects.

pp. 2-21 through 2-23: As I have said in many previous reviews, I believe some mention of the C balance issues as related to N deposition deserves a mention here.

p. 3-7: What about DOE? They have funded a considerable amount of ecological research, including air pollution research.

p. 3-11: Table 3-1 is incomplete. Soils not only provide the service of nutrient cycling, they also provide filters for providing clean water (or, in some cases, provide pollution to clean water). And where are the timber values for forests in here? It is NOT implied only in crops, because there are still forest products removed from National Forests these days, even though their primary purpose is no longer for timbering. The vaguest of all is Climate and especially Climate Control? "Regulating" climate? I did not realize we had that technology yet. What should really be shown here is C balance considerations, but I am aware that the authors are very loathe to do this.

p. 3-25, lines 14-19: Again, potential beneficial effects, for example for C sequestration, are not necessarily limited to managed ecosystems.

p. 3-89 through 3-41: So while potential beneficial effects of N enrichment are summarily dismissed as irrelevant, three full pages are spent on the negative, fire related effects of N enrichment. Certainly the fire effects are very valid ones, that is not the point – it is a matter of selective emphasis on only the negative.

p. 4-2, line 25: Again, acidification of waters can take place in minutes as mobile strong acid anions enter an acidic soil.

p. 4-39, lines 10-16: and D) C sequestration, which can benefit national C balance if permanent and be of benefit, or cause enhanced fire danger in drier systems and thus be extremely negative.

p. 4-45, lines 28-30: This is parsed out very specifically, but is not completely true, as potential benefits can accrue even in unmanaged systems, as stated above.

p. 5-16, lines 4-5 and page 5-7, lines 1-6: These sections clearly point out the problems with steady-state models which assume that base cation flux is equal to soil weathering rate. This simply cannot be true because if it were, soils would never acidify. Acid soils are found all over the world, including in pristine, unpolluted areas.

p. 5-23: CL is not in the list of acronyms

p. 7-7, lines 3-15: This section has me lost. I am unclear as to what is being said here.

p. 7-32, lines 8-13: The real source of uncertainty here is in the assumptions and premises upon which these calculations are based. This source of uncertainty has not been quantified (and perhaps cannot be quantified).

p. A-5, lines 6-11: Nearly all of these assumptions is false. 1. Steady-state conditions never exist, as is well recognized by nearly all ecologists these days. 2. Nutrient cycling effects on soils are profound, often far more important than inputs by deposition and outputs by leaching. 3. N inputs by N-fixation are still greater on a global scale (last time I looked) than those of air pollution (although N fixation is more spotty). 4. I will not contest. 5. Some sesquioxide rich soils can absorb sulfate for a very very long time.

p. A-5, equation 7: This equation and the premises upon which it is based clearly point out the problems with steady-state models which assume that base cation flux is equal to soil weathering rate. This simply cannot be true because if it were, soils would never acidify. Acid soils are found all over the world, including in pristine, unpolluted areas.

p. A-5, line 26: Where is equation 5?

Comments from Dr. Naresh Kumar

Charge question 5: What are the Panel's views on staff's revised conceptual framework for the structure of a multipollutant, ecologically relevant standard for NOx and SOx? To what extent does the Panel agree that this suggested structure adequately represents the scientific linkages between ecological responses, water chemistry, atmospheric deposition, and ambient NOx and SOx?

From a theoretical standpoint the conceptual framework looks fine, but the practical usefulness of the framework depends on its robustness. One way to determine the robustness is by a comprehensive uncertainty analysis, as discussed in my response to Charge Question 14. Another way the AAPI can be tested is by use of historical data. Where data are available, one could use the AAPI (Equation 18 on Page 5-63) to get a trajectory of changes in AAPI in response to changes in SOx and NOy concentrations (I don't know whether concentrations of all forms of SOx and NOy would be available, but some assumptions may have to be made for non-measured components of NOy). The values of other components of AAPI (Q, Neco, [BC]o, LNHx, TNOy and TSOx) have already been estimated by EPA or can be determined using models or measurements. It is critical to do this "hindcasting" at more than one location. The changes in AAPI predicted using Equation 18 should more or less match the changes in ANC (may be with some lag).

Charge Question 6: What are the Panel's views on the appropriateness of considering a single national population of waterbodies in establishing standards to protect against aquatic acidification? What are the Panel's views on consideration of alternative subdivisions of the U.S. to identify the spatial boundaries of populations of waterbodies and acid-sensitivity categories, specifically:

d) the use of Ecoregion III areas to aggregate watrebodies?
e) the use of ANC to further aggregate Ecoregion III areas into different categories of sensitivity?
f) The relative appropriateness of the suggested methods for categorizing spatial boundaries of sensitivity, e.g., one nation, binary sensitive/less-sensitive classes, cluster-analysis based on sensitivity classes, and individual ecoregions?

Using a single national population of water bodies in establishing standards to protect against aquatic acidification may seem attractive for its simplicity, but it has many shortcomings as noted in the PAD and by other members of the panel. Other approaches discussed by the staff are not too complicated either, especially given the overall complexity of the framework, so I see no reason to settle for a single population of water bodies to represent the whole nation. I would recommend using Option 2 (d) unless EPA finds it too complicated in which case 2 (b) or 2 (c) could be used.

Overall, the whole concept of how ecoregions will be used in establishing standards to protect against aquatic acidification was very confusing and EPA needs to do a better job of clarifying this concept.

Charge Question 9: What are the Panel's views on the revised characterization of the deposition transference ratios (TNOy and T SOx)?

A major concern with TNOy and TSOx is that although they are the critical links between NOy and SOx ambient concentrations and their deposition, they are derived using a model that can not be evaluated because of lack of measurements of dry deposition. EPA has shown that inter-annual variability of these ratios can be large even when using the same model. This is most likely due to variability in wet deposition because of changing rainfall patterns from year-to-year. May be EPA should evaluate the stability of the dry and wet deposition ratios separately. It is also critical to show how these ratios vary when using different models and different chemical mechanisms.
It is recommended that EPA evaluate the stability of these ratios by examining these ratios for the following model simulations (in addition to what has already been done):

- CMAQ and CAMx models (it is okay, in fact preferable, to use different emissions and meteorological conditions)
- Different chemical mechanisms
- Different model grid resolutions (36-km v/s 12-km or even 4-km, if available)

Charge Question 10: What are the Panel's views on staff's conclusion that an averaging time of 3 to 5 years is appropriate given the AAPI form of the standard?

The Agency makes a good case for using the averaging time of three years and I agree with that recommendation.

Charge Question 14: What are the Panel's views on the following?

a. The degree to which the chapter appropriately characterizes the potential role of information on uncertainty, sensitivity, and variability in informing the standards?

The discussion of uncertainty and sensitivity analysis is much improved compared to the first draft; however a complete quantitative analysis of uncertainty is needed for the AAPI. Staff has mentioned some good reasons for conducting this analysis, which includes gaining confidence in the data and the models used in defining the form of the standard. The reason it is particularly important to conduct a comprehensive uncertainty and sensitivity analysis for the standard in review is that the many of the components of the AAPI can not be evaluated because of lack of measurements. So, the only way to gain confidence in using AAPI is to examine how sensitive the SOX and NOY response surfaces are to different components of the AAPI. Although, EPA has evaluated uncertainty of some components – some quantitative, but mostly qualitative – the analysis falls short of what the CASAC panel had requested as part of the review of the first draft of the PAD.

The panel had asked the EPA conduct a sensitivity study to characterize uncertainty with different components of the conceptual framework and propagate the resulting uncertainty at every step to arrive at an ensemble of SOX and NOY response surfaces to meet a given level of the AAPI. The range of the SOX and NOY response surfaces would have given a confidence level in the use of the AAPI. By not conducting that level of analysis, EPA has failed to really show how robust an AAPI standard is. It is mentioned in the text that a Monte-Carlo analysis was not performed, but no reason is mentioned for that. I believe it is imperative that EPA conduct a full quantitative analysis of uncertainty before moving forward on using the AAPI construct for setting a standard. One way to conduct the quantitative uncertainty analysis is to use the Monte Carlo approach to arrive at an ensemble of SO_x and NO_y response surfaces to meet a given AAPI standard. As an example, the Monte Carlo analysis for Adirondacks could be conducted by using the following approach:

- Use a probable range (or a mean value with some assumed or derived standard deviation) of estimates for Neco,
- Use a probable range (or a mean value with some assumed or derived standard deviation) of estimates for $[B_C]_0$,
- Use an estimate of Q based on different years (dry vs. wet year),
- Calculate $L(NH_x)$, T_{NOy} and T_{SOx} using different air quality model simulations that may already be available and use the distributions in Monte Carlo simulations,
    - Use CMAQ, CAMx, or other model simulations for different years (with different meteorological conditions and different emissions) to get a range of these variables,
    - Use different chemical mechanisms, if available
- Use covariant constraints for quantities that may be correlated when running the Monte Carlo simulations

b. The appropriateness and completeness of the evaluation of CMAQ model performance and sensitivity to critical inputs?

The performance of the CMAQ model is still incomplete after repeated requests by the CASAC panel. The performance statistics for the major species shown in Table 7-1 can be misleading. One should use mean normalized bias (and not normalized mean bias) when conducting model performance using concentrations that are averaged over long periods (e.g. one week or more). A complete evaluation of CMAQ derived values with measurements is needed before any confidence can be placed on the use of the model to generate the desired parameters. Although the model cannot be evaluated for dry deposition because of lack of measurements, evaluation of the ability of the model to represent ambient levels can serve as a proxy for its ability to represent dry deposition. It is recommended that following evaluations (using daily or weekly averaged quantities, not annual) be performed to assess the uncertainty in the model:

4. Model performance for nitric oxide, nitrogen dioxide, sulfur dioxide, nitrate, ammonium and aerosol nitrate, ammonium, and sulfate for different networks for which the data are routinely available,
5. Model performance for wet deposition of sulfate, nitrate, and ammonium using the National Atmospheric Deposition Program (NADP) network,
6. A regional model evaluation using the continuous measurements of nitric oxide, nitrogen dioxide, nitric acid and NO_Y from the SEARCH network in the southeastern U.S.

c. The utility of the analyses of temporal and spatial variability in the deposition transference ratios (TNOyand TSOx)?

Showing year-to-year change using the same model is not sufficient. A complete analysis should also show how these ratios vary with use of different models and different chemical mechanisms. EPA has the modeling data from the CAMx model used for the transport rule, so it should be able to compare these ratios easily from model-to-model.

Charge Question 15: What are the Panel's views on the insights provided by the AAPI sensitivity analysis including: a. The evaluation of elasticities of response? b. The multivariable ANOVA analysis?

Evaluation of elasticity of response is a good way to get a first hand picture of the sensitivity of the AAPI wrt to different components of the AAPI. However, I would suggest doing this analysis for the SOX and NOX concentrations needed to meet a standard, as those are the quantities for which the standard is being set.

Charge Question 24. In light of the Panel's views on what constitutes adverse effects to public welfare (see Chapter 3), what are the Panel's views on:

b) target values for ANC that protect against adversity to public welfare in light of the information presented in Chapter 5 concerning levels of ANC and the ecosystem effects associated with those target ANC levels?

d) alternative standards for NOx and SOx that would protect against adverse effects to public welfare based on the AAPI form, and taking into account
(i) consideration of target levels of ANC (chapter 5),

Given the lack of complete quantitative uncertainty analysis, I don't think the administrator would have a high degree of confidence in whether a particular level of AAPI would indeed provide requisite level of protection or be overly protective for that matter. Qualitative way the uncertainty has been discussed has the potential to give a false sense of confidence about the overall uncertainty. On the bottom of Page 7-2 and top of Page 7-3, there is a discussion on what a low level of confidence in the components of the standard mean. It goes on to say that if the confidence is low then

AAPI could be adjusted upwards or downwards depending on whether you want to put more emphasis on providing requisite level of protection or on not making the standard being overly protective. I would add one more thing there is that if the level of confidence is too low, then it makes the AAPI form of the standard questionable whether it is a robust standard or not. The reason it is particularly important to conduct a comprehensive uncertainty and sensitivity analysis for the standard in review is that many of the components of the AAPI can not be evaluated because of lack of measurements. So, the only way to gain confidence in using AAPI is to examine quantitatively how sensitive the SOX and NOY response surfaces are to different components of the AAPI. Given the lack of a quantitative sensitivity analysis we have no idea of the confidence interval and in my view it is premature to talk of the levels or ranges of standards unless that shortcoming is overcome.

Comments from Dr. Myron Mitchell

Three important areas that need to be addressed are:

1) Evaluation of AAPI using historical data will be very helpful in developing this combined NOy and SOx standard. There are a number of historical data sets including the Adirondacks, Catskills, Hubbard Brook, etc. that have long-term water chemistry data. There may be some difficulties in obtaining all of the needed AAPI inputs including atmospheric deposition estimates, but at least some approximation will be very useful. This use of historical data will provide additional confidence in the AAPI approach.

2) The reliance on the CMAQ model with respect to providing estimates of deposition input is important to clearly link this effort by EPA with effects. The importance of the CMAQ output for developing this secondary standard clearly suggests that more effort is needed by EPA in the evaluation of the CMAQ output. This should evaluation should be a high priority for EPA in monitoring and research efforts.

3) Treatment of sulfur in the AAPI. There is no consideration in the AAPI formulation of internal (soil) sulfur sinks (e.g., soil sulfate adsorption) or sulfur sources (organic S mineralization, S mineral weathering, sulfate desorption). It is assumed that watershed sulfur outputs equal sulfur inputs in deposition. Mitchell[1] et al. (2010, Biogeochemistry) found that watersheds that had previously had substantial portions of atmospheric S input that from 1985 through 2002 that internal sources contribute 1–6 kg S ha^{-1} $year^{-1}$. This would equal 6 to 37 $meq/m^2/year$. This contribution is substantial when compared to various analyses provided in the document (e.g., figures 5-15, 5-18, 5-19, etc.). Not including this internal sulfur source will result in an underestimate in the amount of reduction for nitrogen and sulfur deposition needed to meet target loads. Other studies in North America and Europe have also emphasized the importance of internal sulfur sources.

My other comments are given below.

Executive Summary

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[1]Mitchell, M.J., G. Lovett, S. Bailey, F. Beall, D. Burns, D. Buso, T. A. Clair, F. Courchesne, L. Duchesne, C. Eimers, D.Jeffries, S. Kahl,, G. Likens, M.D. Moran, C. Rogers, D. Schwede, J. Shanley, K. Weathers and R. Vet. 2010. Comparisons of Watershed Sulfur Budgets in Southeast Canada and Northeast US: New Approaches and Implications. Biogeochemistry (In Press and Available on Line).

Page(s)

ES-8 The term Neco used in the figure has not been defined prior to its use.

ES-9 The sentence “Snowmelt can release stored N deposited throughout the winter” is conceptually not correct. The vast majority of N released is nitrate that has been generated microbially within the soil, not atmospherically deposited N in the snowpack.

Chapter 1: Introduction
[no questions]

Chapter 2: Known or Anticipated Ecological Effects
[no questions]

Chapter 3: Considerations of Adversity to Public Welfare

3-15 to16 It would be useful to indicate in figure legends 3-5 and 3-6 that it is assumed that all the N and S deposited are converted to nitrate and sulfate respectively for calculation eq/ha/yr.

3-28 Table 3-2 needs further clarification. It is not completely clear why the lake count is 0 for years 2005 for all ANC thresholds.

3-29 For Table 3-3 include within the table legend a replacement of “present” with “Year 2007".

3-37 In Table 3-7, a delineation of the arrows used in the value column needs to be provided.

1. What are the Panel’s views on the definitions of adversity that are appropriate to consider in determining what constitutes adversity to public welfare relative to the NOx and SOx secondary standards?

The Chapter does a good job of describing the various attributes of diversity with particular emphasis on those areas expected to be most sensitive to NOx and SOx effects in the USA. The impact of the Chapter could be improved by a summary section that clearly indicates which of the adversity components will be the primary focus of the proposed standards.

Chapter 4: Addressing the Adequacy of the Current Standards

4-5 line 4 This sentence needs to be changed from “oxidized nitrogen” to "reactive nitrogen"–as it currently is written it excludes reduced forms of N including ammonium.

4-15 In describing the issues related to the differences between the rural (e.g., CASTNET) and urban deposition monitoring sites, it is clear that there is a disconnect. Would it be appropriate to recommend that a unified network is needed that includes both rural and urban sites?

4-17 to18 Certainly there is justification for using CMAQ as a predictor of deposition. It is somewhat curious, however, that NADP is used for wet deposition and CMAQ is for dry deposition. Certainly, there are more problems associated with the estimates of dry deposition than those for wet deposition. However, to gain more confidence in the CMAQ predictions it would be very important to compare the NADP (measurements) and CMAQ (predictions) for wet deposition. This type of comparison is needed to confirm that "CMAQ promotes analytical consistency and efficiency across analyses of multiple pollutants" and "CMAQ provides a consistent platform incorporating the atmospheric and deposition species of interest over the entire United States".

4-18 The issues related to scaling up in time the CMAQ estimates of hourly estimates needs to be discussed.

4-21 to 36 It would be very helpful to use the same color ranges for each gases pollutants for comparing estimates from CMAQ, CASTNET and SLAMS. Also, there is very limited discussion on the differences in the results associated with these different monitoring networks. For example, there appear to be major differences in CMAQ (Figure 4-11) and CASTNET (Figure 4-13) sulfate concentrations with respect to the absolute values and spatial distribution.

2. What are the Panel's views on staff's approach to translating the available evidence and risk information and other relevant information into the basis for reaching conclusions on the adequacy of the current standards and on alternative standards for consideration?

The general information is certainly contained within this document and other supporting information such as within the ISA and REA, but the actual linkages of evidence and the translation to the generation of the standards could be improved.

a) In light of the Panel's views on the appropriate definitions of adversity to public welfare (see Chapter 3), do you agree that the current levels of NOy and SOx deposition are adverse to public welfare?

b)

Yes, the evidence is sufficient that the current levels of NOy and SOx deposition are adverse to public welfare in some systems which are particular sensitive to acidification and or N addition causing nutrient enrichment.

3. Has staff appropriately applied this approach in reviewing the adequacy of the current standards and potential alternative standards?

Yes, the approach is valid, but more attention to the linkages between evidence and the generation of the standards would be helpful.

4. Has staff appropriately acknowledged the potential beneficial effects of nitrogen inputs into nutrient limited ecosystems, while maintaining the focus of the review on preventing adverse effects in nitrogen sensitive ecosystems?

The current balance is appropriate in the context of the standard and the protection of sensitive systems.

Chapter 5: Conceptual Design of an Ecologically Relevant Multi-pollutant Standard

5-5 lines 5-6 This statement is not correct. The vast majority (>95%) of the nitrate released during episodic snowmelt is derived from the forest floor and mineral soil and not from the snow. A possible rewording could be as follows: Snowmelt results in the mobilization to drainage waters of nitrate most of which has been generated within the forest floor and mineral soil. This release of this nitrate can result in episodic acidification. Literature citations would include (Kendall, 1998, Tracing Nitrogen Sources and Cycling in Catchments, Book Chapter; Piatek et al., 2005, WASP; Campbell et al. 2006, J. Geophys. Res.).

5-5 lines 7-8 The statement that “inputs of nitrogen and sulfur from snowpack and atmospheric deposition” suggests that snowpack N and S is not derived from atmospheric deposition. Change to “inputs of nitrogen and sulfur from atmospheric deposition”.

5-12 18 Lien et al. 1992 not in References.

5-28 lines 10-22 This section is difficult to follow. Inclusion of a figure illustrating the issue associated with the skewness of the distribution of critical loads would be helpful so that the reader does not need to go ahead to section 5.3.2.7 to understand the issue.

5-33 Figure 5-9 is very difficult to read. The numerical designations of ecoregions especially in the dark blue areas are not readable.

5-45	Figure 5-13. In its present form it is difficult to distinguish differences between the one nation versus binary categorization.
5-55	lines 26-29 The statement "Due to lack of direct measurements, no performance evaluations of CMAQ's dry deposition calculation can be found; however, the current state of MCIP is the product of research that has been based on peer-reviewed literature from the past two decades (EPA, 1999) and is considered to be EPA's best estimate of dry deposition values" is rather weak and suggests that effort is needed to further evaluate CMAQ using available information. This issue comes up a number of times in the document (e.g., page 5-64, lines 23-25).
5-56	lines 22-28 The time unit for these depositions and ratios needs to be provided. Is this a yearly interval?
5-57	In showing these coefficient of variation values in Figure 5.22, it is difficult to see the actual values and respective ranges in the Adirondack and Shenandoah case study areas. Instead of stating that "values are relatively small", it would be better to provide the means and standard error of the means of these ratios.
5-58	In Figure 5-20, for sulfur deposition/concentration, how is marine sulfur accounted for? For sulfur it seems somewhat curious that there is a change in the isopleths a substantial distance into the Atlantic Ocean. I would expect the difference if it includes marine components would be more related to the coastal outline. For this figure, the deposition component needs a time unit as previously stated.
5-61 to 70	I am concerned with the treatment of sulfur in the AAPI. There is no consideration in the formulation of sulfur sinks (e.g., soil sulfate adsorption) or sulfur sources (organic S mineralization, S mineral weathering, sulfate desorption). Mitchell et al. (2010, Biogeochemistry) found that watersheds that had previously had substantial portions of atmospheric S input that from 1985 through 2002 that internal sources contribute 1–6 kg S ha^{-1} $year^{-1}$. This would equal 6 to 37 $meq/m^2/year$. This contribution is substantial when compared to various analyses provided in the document (e.g., figures 5-15, 5-18, 5-19, etc.)
5-84	lines 12-23 In considering issues related to recovery there is a need to not only consider the issues related to weathering of base cations, but also to internal generation of the mobile nitrate and sulfate anions. Particularly for sulfate this sulfate will likely result in substantial delays in recovery in those systems with net losses of soil sulfur and low levels of base cation weathering.

5. What are the Panel's views on staff's revised conceptual framework for the structure of a multipollutant, ecologically relevant standard for NOx and SOx? To what extent does the Panel agree that this suggested structure adequately represents the scientific linkages between ecological responses, water chemistry, atmospheric deposition, and ambient NOx and SOx?

> The conceptual framework for a multipollutant, ecologically relevant standard for NOx and SOx is sound with considerable support from the scientific literature on how the generation of strong mobile acids result in the acidification of soils and water. Some of information, however, is not correct or incomplete. Note for example the discussion of nitrate sources during snow melt as discussed above for page 5-5, lines 5-6. Also the assumptions associated with atmospheric sulfur input being equal to drainage water losses are also not correct (see page 5-61 to 70).

6. What are the Panel's views on the appropriateness of considering a single national population of waterbodies in establishing standards to protect against aquatic acidification?

> Although having a single national population of waterbodies makes is more facile to explain the standard, the problems associated with under protecting sensitive systems and overprotecting insensitive systems necessitates having a system with more spatial resolution.

What are the Panel's views on consideration of alternative subdivisions of the U.S. to identify the spatial boundaries of populations of waterbodies and acid-sensitivity categories, specifically:

a) the use of Ecoregion III areas to aggregate waterbodies ?

> This seems to be a reasonable approach that takes advantage of the extensive information on various ecosystem components including both abiotic and biotic components.

b) the use of ANC to further aggregate Ecoregion III areas into different categories of sensitivity?

> The use of ANC is consistent with the overall emphasis on the standard to protect sensitive surface waters from further acidification and have deposition that will allow those water bodies that have been deleterious impacted by acidic deposition to recover as indicated by increasing ANC values.

c) the relative appropriateness of the suggested methods for categorizing spatial boundaries of sensitivity, e.g. on nation, binary sensitive/less-sensitive classes, cluster analysis based sensitivity classes, and individual ecoregions?

The analysis is interesting showing the different distributions of sensitivity and that categorization that captures substantial variation should be used. This will be a compromise between one nation versus using individual ecoregions.

7. What are the Panel's views on the appropriateness of the critical loads that form the basis for the population assessment to determined deposition metrics?

The use of critical loads has been found to be a useful approach for looking at spatial and temporal aspects of acidification. This concept was originally applied to Europe and more recently has been extended to other regions including North America.

a) What are the views of the Panel on the appropriateness of generalizing the f-factor approach to apply to lakes and streams in the Western U.S. and other portions of the Eastern U.S.

The application of the f-factor is a useful approach for evaluating the potential for mineral weathering to contribute to the generation of base cations and enhance acid neutralization.

b) What are the views of the Panel on the filtering criteria used to remove lakes and streams that are naturally acidic or not sensitive to atmospheric deposition?

Yes, it is reasonable to exclude lakes in two of these classes: 1) CL < 10 meq/m^2/yr and for which pre-industrial ANC values could not be calculated; and 2) waters affected by acid mine drainage (>400 μeq/L SO_4^{2-} twice or more than expected by atmospheric deposition. Lakes with low ANC values (e.g. < 50 μeq/L) and waters with >10 mg C/L DOC those dominated by organic acids should be included in the analyses and possibly flagged with respect to these characteristics. Alternatively, the PA should show the consequences related to excluding or including these with respect to the development of the AAPI and critical loads.

8. What are the Panel's views on the suggested methods for determining appropriate values of reduced nitrogen deposition in establishing NOx/SOx tradeoff curves?

The presentation of the NOx/SOx tradeoff curves is difficult to follow since the linkages between the various components in the various figures and tables are not always consistent (e.g. Table 5-7 versus Figure 5-12). Also it would be most helpful to keep the axes lengths the same in plots within the same figures for comparisons (e.g. Figure 5-15). I am somewhat concerned with respect to the sulfate portion of the curve on how systems are handled in which sulfate losses in drainage waters is not in balance with sulfur deposition. We know that for a number of sites in the United States that there can either be net retention or net loss of sulfur. These imbalances can be substantial especially under conditions of decreasing atmospheric sulfur deposition.

9. What are the Panel's views on the revised characterization of the deposition transference ratios (TNOy and TSOx)?

> Implicit in the use of such an aggregated deposition ratio is that the relative portion of the chemical species in deposition remain constant both with space and time. This analysis was done for only from 2002 – 2005 and hence may have covered a range of meteorological conditions, but little difference in nitrogen and sulfur sources. I don't believe that there is strong evidence suggesting that this is the case. At a minimum some error analyses associated with this assumption is needed. This analysis should be expanded beyond the results of CMAQ and to include other estimates of the proportion of gaseous species and their respective deposition velocities. It is recognized that this other analysis will include fewer chemical species than that provided in CMAQ.

10. What are the Panel's views on staff's conclusion that an averaging time of 3 to 5 years is
appropriate given the AAPI form of the standard?

> There should be consideration of not only looking at the averages, but also the minima and maxima for the period of examination. Care will need to be taken on issues related to the water regimes and other climatic factors among these years. Droughts or other extreme events such as freezing rain can have a substantial impact on N and S drainage losses and resultant effects on ANC.

11. What are the Panel's views on the preliminary staff conclusions regarding alternative target ANC levels that are appropriate for consideration and the rationale upon which those conclusions are based?

> The use of alternative ANC levels is appropriate and based upon sound science that has shown different levels of sensitivity of various biotic taxa with respect to sensitivity to low ANC.

a) In light of the Panel's views on the appropriate definitions of adversity to public welfare (see Chapter 3), what are the Panel's views on the appropriateness of the information related to adversity considered by staff in evaluating alternative target ANC levels?

> The information provided is adequate for showing at least some of the major concerns that are documented with respect to public welfare.

12. What are the Panel's views on the approaches considered by staff for assessing alternative target percentages of water bodies for protection at alternative ANC levels?

> This approach is useful in providing a range of water bodies to be covered with respect to these alternative ANC levels. This also provides flexibility with

respect to the administrator regarding choices for protection, overall protection of public welfare and costs for implementation of the standard.

Chapter 6: Co-protection for Other Effects Using Standards to Protect Against Aquatic Acidification

13. What are the Panel's views on the utility of the additional analyses of co-protection benefits to inform consideration of alternative levels of the standard?

This discussion is helpful in showing the linkage of protection between terrestrial and aquatic components of watersheds and emphasizes as indicated elsewhere in the document that in general the projection of sensitive aquatic resources results in terrestrial protection with the aquatic resources being more sensitive to deposition. One issue, however, that needs some consideration is that the time frames for the recovery are substantially different from aquatic and terrestrial components with a greater time lag expected for terrestrial systems.

Chapter 7: Evaluation of Uncertainty and Variability in the Context of an AAPI standard, including Model Evaluation, Sensitivity Analyses, and Assessment of Information Gaps

14. What are the Panel's views on the following:

a. The degree to which the chapter appropriately characterizes the potential role of information on uncertainty, sensitivity, and variability in informing the standards?

7-4 lines 13-15 The document states that "Confidence regarding the fundamental science supporting causal determination about the effects of acid deposition, and the translation of those efforts into ecosystem services and values is less amenable to quantification". Even though it is difficult, providing even some approximate evaluations would be helpful. Some of this uncertainty may be substantial and having uncertainly analysis focus on those components for which the calculations are more facile may result in a misunderstanding of the impact of the proposed standard on human welfare.

7-6 lines 14-17 In addition to the uncertainties associated with the estimate of catchment supply of base cations via weathering the exclusion of sulfate dynamics (or possibly considering a range of internal S supply) will have a major impact on uncertainty especially associated with future recovery.

7-8 lines 13-23 Some further elaboration of the Banzhaf survey would be helpful.

b. The appropriateness and completeness of the evaluation of CMAQ model performance and sensitivity to critical inputs?

7-10 to 24 The inclusion of comparisons of CMAQ and CASTNET results (i.e., Figures 7-1 to 7-7) is very helpful. The discussion related to the limitations of CMAQ (page 7-10, lines 15-26) are insightful and should be useful in providing future modifications of CMAQ.

7-12 line 1 Is it appropriate to utilize a manuscript in preparation (e.g., Dennis and Foley, 2010) for this document?

7-3 lines 12-27 It is indicated that "sensitivity of CMAQ derived deposition transformation ratios to changes in emissions and treatment of chemistry" is not yet completed. This should be a high priority for EPA.

7-12 lines 1-7 Do these results suggest that CMAQ needs to be changed such that precipitation estimates are derived from actual measurements versus modeled estimates. Isn't this approach more similar to that employed by the Canadian AURAMS model?

c. The utility of the analyses of temporal and spatial variability in the deposition transference ratios (TNOy and TSOx)?

7-13 The terms "stiff" and "stiffness" are introduced. Is the use of these terms identical to "invariate"? In indicating that the absolute values remain "stable", it is difficult to ascertain how these relative large ranges of ratios will affect the overall results in using mean Ts and Tn values.

7-14 This comparison to emission change over time is for only two years (2005 and (2030) and is highly dependent on assumptions of changes in emission sources. What were the underlying assumptions of these changes? Do the range in values in Figure 7-12 show the differences based upon these assumptions?

7-16 lines 14-26 With the continual evolution of CMAQ and likely changes in the predictions of AAPI, will there be problems in the standard itself being affected by changes in CMAQ?

7-27 to 28 For Figures 7-11 and 7-12, the figure legend needs to include a description of the statistical values (mean, ranges, confidence intervals, ??) associated with these box-and-whisker plots.

15. What are the Panel's views on the insights provided by the AAPI sensitivity analysis including: a. The evaluation of elasticities of response?

b. The multivariable ANOVA analysis?

7-30 It is challenging to use the results provided in Appendix A and see how the various analyses are used in this evaluation. A summarization is needed on the relative sensitivities of the various parameters that make up the AAPI. An important result is provided by the statement that emphasizes the need to focus on the uncertainties of the non-atmospheric inputs, including base cation weathering and runoff rates. As indicated previously some inclusion of internal generation of sulfate is also needed. An important outcome of this analysis should to show the parameters of the AAPI have the most and least confidence. Such information should be used in driving research and monitoring efforts by EPA.

16. What are the Panel's views on the discussion of uncertainty in the critical loads models including MAGIC and SSWC?

These descriptions of uncertainty for the model calculations for MAGIC and SSWC are adequate. A more quantitative term than "moderate' should be used in describing the uncertainty in SSWC (Page 7-32, line 15). The development of the information in Table 7-2 is helpful in summarizing the uncertainty associated with the AAPI.

7-33 to 34 For Figures 7-14 and 7-15 within the figure legend it needs to be indicated that these MAGIC model simulations.

17. What are the Panel's views on the areas for future research and data collection outlined in this chapter, on relative priorities for research in these areas, and on any other areas that ought to be identified?

The AAPI needs to include some estimates of the role of internal sulfur sources in contributing to sulfate in drainage waters. The absence of including this factor will result in an underestimate of the deposition required to achieve a desired level of ANC.

Chapter 8: Monitoring

8-1 to 18 The most critical aspect of monitoring is that there needs to be a more explicit linkage between the monitoring networks and the evaluation and further refinement of the CMAQ model. This interplay between the monitoring and modeling efforts will help ensure that both the monitoring and modeling are most relevant to the environmental issues being addressed.

18. What are the Panel's views on using an open inlet to capture all particulate size fractions for the purpose of analyzing for sulfate?

This should be the focus of a research question versus an overall modeling component.

What is your opinion on using existing CASTNET filter packs as a future Federal reference method for sulfate?

> This has considerable advantages with respect to spatial and temporal patterns since the CASTNET network has been in place for a number of years and includes a generally good representation of sites across the US.

19. What are the Panel's views on requiring measurements of ammonia and ammonium to assist implementation of the standard?

> There is a clear need to expand monitoring to include measurements of ambient ammonium and ammonium concentrations. This reduced form of nitrogen is a major component of nitrogen deposition for many sites including those within areas with intensive agricultural activities.

20. What are the Panel's views on having a subset (e.g., 3-5 sites) of monitoring stations in different airsheds that measure for the major NOy species; nitric acid, true NO2, NO, PAN and p-NO3?

> This could be an important research question.

Chapter 9: Conclusions

21. What are the Panel's views on the overall characterization of uncertainty as it relates to the determination of an ecologically-relevant multi-pollutant standard for NOx and SOx?

> The current document does a commendable job in showing were some of the major uncertainly lies with respect to the development of a multi-pollutant standard. Areas that should be targeted for improvement include a more complete evaluation of the CMAQ predictions and the consideration of additional processes, especially internal sulfur sources in the AAPI.

22. What are the Panel's views on the following:

> a. The insights that can be gained into potential alternative additional secondary standards (using the AAPI form) by considering:
>
> > I. Information from studies on the relationship between mortality in aquatic organisms and pH and ANC?
> > ii. Information from studies on the relationship between fish health and/or biodiversity metrics and pH and ANC?
> > iii. Information on the relationship between pH, Al, and ANC?
> > iv. Information on target ANC levels identified by states and regions, as well as other nations?
> >
> > Each of these sources of information both separately and taken together

provide a compelling case on the relationships between ANC and other water quality metrics that are associated with biotic health of waters. The findings from these different studies all provide a rather unified picture suggesting appropriate ANC values to be the target for the standard.

b. The appropriate role of qualitative and quantitative characterizations of uncertainty in developing standards using the AAPI form?

As mentioned previously the uncertainty of the AAPI needs to include not only statistical analyses associated with specific model parameters, but also include an evaluation of possible omissions in the AAPI (e.g. reduced nitrogen inputs, contribution of sulfate sources and sinks in soil, etc).

c. The role of considerations regarding the relationship of the standard to:

i. the time trajectory of response, e.g. when specific ANC levels are likely to be realized given a specific level of the AAPI?
ii. the likelihood of damages to aquatic ecosystems due to episodic acidification events given a specific target for chronic ANC?
iii. the levels of co-protection for terrestrial ecosystems against acidification effects and the for aquatic and terrestrial ecosystems against effects of excess nutrient enrichment?

There may be some problems associated with the time trajectory of the response due to the understanding and ability to model the relative contribution of net N uptake and net S loss from the terrestrial portion of the system. Any factor (e.g. changes in climate, CO_2 concentration in the atmosphere) could have important effects on the time trajectory. Also, the effect of factors may be substantially different between aquatic and terrestrial ecosystems. For example, although for aquatic systems the sum of base cations may be adequate, for terrestrial systems the availability of specific base cation, calcium, may be a critical factor in affecting tree health. Important tree species such as sugar maple have a high demand for calcium.

23. What are the Panel's views on Staff's conclusion that the existing secondary standards for NOx and SOx should be retained to provide protection against direct adverse effects to vegetation due to gas phase exposures?

There is no reason not to retain these existing standards since these concentration levels will likely be substantially greater than those associated with join NOx and SOx standards. More importantly the scientific justification is still valid for protecting against deleterious impacts to vegetation.

24. In light of the Panel's views on what constitutes adverse effects to public welfare (see Chapter 3), what are the Panel's views on:

a) the degree to which current levels of NOy and SOx deposition are adverse to public welfare based on evidence and risk information, and information on adversity provided in Chapters 2,3, and 4?

b) target values for ANC that protect against adversity to public welfare in light of the information presented in Chapter 5 concerning levels of ANC and the ecosystem effects associated with those target ANC levels?

c) factors relevant in selecting target percentages of waterbodies to protect at alternative target ANC levels to protect against adverse effects to public welfare, and weights to place on those factors?

The information provided substantiates that the current levels of NOy and SOx deposition are producing adverse effects to the public welfare. The target values selected for ANC are congruent with current scientific understanding with respect to which ANC values and any resultant change are most sensitive to biotic components. Selecting a target subset of waterbodies to be protected by alternative target ANC values is a useful approach.

d) alternative standards for NOx and SOx that would protect against adverse effects to public welfare based on the AAPI form, and taking into account

- consideration of target levels of ANC (chapter 5),
- target percentage of water bodies to protect (chapter 5),
- consideration of relevant uncertainties in AAPI components (chapter 7), and
- any other potentially relevant factors, such as levels of co-protection against terrestrial acidification and nutrient enrichment (chapter 6)?

It may be important to consider alternate standards especially for protecting those systems where nutrient enrichment (e.g. western U.S.) is a substantial effect associated with N deposition.

Comments from Mr. Richard Poirot

5. What are the Panel's views on staff's revised conceptual framework for the structure of a multipollutant, ecologically relevant standard for NOx and SOx?

The revised conceptual framework for the structure of the multi-pollutant secondary standard has been substantially improved from the first draft policy assessment. The inherently complex framework is more clearly presented and more carefully justified, with revisions that are directly responsive to previous review comments.

To what extent does the Panel agree that this suggested structure adequately represents the scientific linkages between ecological responses, water chemistry, atmospheric deposition, and ambient NOx and SOx?

The proposed structure adequately reflects the current state of scientific understanding of the complex linkages between ambient concentrations of SOx and NOx, wet and dry deposition of these and other acidifying pollutants (i.e. NHx), environmental processing of these deposited S and N compounds, resultant changes in surface water chemistry, and subsequent ecological effects.

6. What are the Panel's views on the appropriateness of considering a single national population of waterbodies in establishing standards to protect against aquatic acidification?

Use of a single national population of water bodies as the basis for selecting (a percent of water bodies to be protected from reaching or maintaining a specific minimal ANC component of) a national standard has the "advantage" of "simplicity". But a large fraction of national surface waters are located in areas where underlying soils, bedrock and other local environmental factors effectively preclude adverse acidification effects from past, current, and expected future deposition rates of S and N, while other water bodies are extremely sensitive to effects from relatively low rates of S and N deposition. Use of a single national population and associated percentage level of protection unnecessarily disregards the large regional variations in inherent sensitivity to acidification, and is likely to lead to under-protection in some areas and over-protection in others (or both).

Since there are various methods and data available to allow refined estimates of inherent sensitivity to be calculated on a regional basis, and since many other location-specific environmental variables are included in the calculation of compliance with the proposed standard, it makes sense (I think – but need more info) to consider protection in the more refined context of the populations of water bodies at risk from acidification effects.

What are the Panel's views on consideration of alternative subdivisions of the U.S. to identify the spatial boundaries of populations of waterbodies and acid-sensitivity categories, specifically:

a) the use of Ecoregion III areas to aggregate waterbodies ?

Not my area of expertise, but this seems like a reasonable approach, and possibly one that could be considered for refining secondary NAAQS for these and other criteria pollutants in the future. Off hand, it seems like using 120+ different Ecoregion III categories for aggregating water bodies is unnecessarily complex. However, it also appears that there are reasonably ways to simplify, group or sort these many categories into a much smaller number of Ecoregion subsets which are inherently sensitive (or most sensitive) to acidification, and which would make for a more efficient standard better focused on protecting those systems at greatest risk.

b) the use of ANC to further aggregate Ecoregion III areas into different categories of sensitivity?

This seems like a logical (almost obvious) metric for sorting/grouping the Ecoregion categories. If only we could just use the readily measured direct ANC indicator of effects as the NAAQS indicator…

c) the relative appropriateness of the suggested methods for categorizing spatial boundaries of sensitivity, e.g. on nation, binary sensitive/less-sensitive classes, cluster analysis based sensitivity classes, and individual ecoregions?

I don't have a strong opinion on the relative appropriateness of these alternative approaches. None of them seems inappropriate. Off hand, I think I like the cluster analysis approach, for its inherent scientific merit, its direct focus on sensitivity, and the relative simplicity of a 5-class grouping scheme (especially for the initial roll-out of an extremely complex NAAQS).

However, I also don't think that the advantages/disadvantages/ consequences of the various options (2a, b, c, d) are presented here with sufficient clarity to allow an informed choice by the Administrator (or by me anyway). Hopefully, these options can be presented, discussed and illustrated more clearly in the final PA document, and staff might propose and defend a preferred option. For example, the page 5-50 statement (and associated figures) that "In option I, the Adirondack air quality is slightly out of attainment for a 75%-tile deposition metric based on a CL at ANC=50. In option 2a, the Adirondack air quality is out of attainment for the curve for the sensitive areas, but in attainment for the less sensitive areas." helps convince me that option 2a is preferable to option 1. But I don't have a similar feel for the relative strengths or weaknesses of the other options.

8. What are the Panel's views on the suggested methods for determining appropriate values of reduced nitrogen deposition in establishing NOx/SOx tradeoff curves?

Since reduced nitrogen in the air or in (dry) deposition is not currently measured, and not currently considered as a regulated component of the NAAQS, but does contribute to the acidifying (and N enrichment) effects of SOx and NOx deposition, I think it's reasonable to estimate its location-specific deposition with CMAQ. At the same time, there is also a need to verify and refine the CMAQ estimates with direct measurements, especially for NH_3.

I also strongly support the proposed approach to consider NHx as a temporally varying, location-specific component of the AAPI calculation. This is a scientifically preferable approach to the previous proposal which would have considered NHx deposition as a fixed constant. If NHx increases, larger reductions in SOx and NOx would be required, and conversely, if NHx decreases, SOx and NOx reductions would be lower.

9. What are the Panel's views on the revised characterization of the deposition transference ratios (TNOy and TSOx)?

So far as I can tell, the (CMAQ) methods for calculating these deposition transference ratios are the same as they were in the last draft PA, but are described, illustrated and evaluated more clearly in the current document. These transfer functions are logically conceived, but seem like such critical elements of the proposed standards, which are uncomfortably dependent entirely on CMAQ model performance. The illustrations (Figures 5-20 – 5-22) showing the spatial characteristics are helpful, and the illustrations (Figures 7-11 and 7-12) showing that the transfer functions remain stable over time with large changes in emissions add some confidence. Although since the model chemistry is fixed, S and N species totals are conserved, and meteorology held constant, highly variable modeled results would not be expected. Some additional confidence might be provided by comparing CMAQ estimates of dry and total deposition (wet is already shown) at selected CASTNET sites in recent years, and perhaps breaking out the Figure 7-3 performance for TNO_3 into separate figures for particulate nitrate (which deposits inefficiently) and HNO_3.

One additional analysis that might be informative would be to calculate and evaluate a modified T_{NOY} function (call it T_{NOY*}) that would be based on CMAQ modeled total N deposition as a joint function of CMAQ HNO_3 and pNO_3 (separate coefficients could be derived for each species). This empirically derived relationship would be no more of a "black box fudge factor" than the current T_{NOY} calculation (ratio of CMAQ estimate of total N deposition to CMAQ estimate of NOy). Potential benefits of this approach are that it would be less dependent on CMAQ's ability to accurately predict and apportion all the separate NOy components (with their widely different deposition velocities); it can be applied (as can the sulfur T_{SOX} function) to currently available and relatively low cost CASTNET measurement data; and the measured species would directly represent major components of dry N deposition, compared to NOy, which has no relationship ($R = 0.067$ in Figure 4-21) without benefit of the black box CMAQ conversion. In evaluating whether this alternative approach is "close enough" to the original TNOY, both calculations could be compared to both the CMAQ estimates and CASTNET (+ NADP) measurements of total N deposition.

12. What are the Panel's views on the approaches considered by staff for assessing alternative target percentages of water bodies for protection at alternative ANC levels?

As indicated previously, I think the alternative approaches seem reasonable, and that the objective should be to focus as tightly as possible on protecting water bodies that are inherently sensitive to acidification, without adding too much complexity to the regulatory metric. It might also be recognized in this case that a metric that provided some "over-protection" in areas less sensitive to aquatic acidification might provide added protection for terrestrial ecosystems or against nitrification and would unavoidably improve visibility and reduce mortality and morbidity associated with $PM_{2.5}$ concentrations.

13. What are the Panel's views on the utility of the additional analyses of co-protection benefits to inform consideration of alternative levels of the standard?

This seems like a reasonable concept to explore in more detail, although I don't really see any discussion of this in Chapter 6. It seems clear from the analysis that there are areas – without surface waters or with relatively insensitive surface waters where adverse affects on terrestrial ecosystems are expected – and for which adding a "co-protection" element to the standard would provide added benefits. If other welfare effects of SOx and NOx – such as on materials damage and visibility had been considered in this review, the co-protection benefits would have been substantial.

14. What are the Panel's views on the following:

a. The degree to which the chapter appropriately characterizes the potential role of information on uncertainty, sensitivity, and variability in informing the standards?

The additional information and discussion uncertainty, sensitivity, and variability in Chapter 7 is extremely helpful, and represents a major improvement to the previous draft PA.

b. The appropriateness and completeness of the evaluation of CMAQ model performance and sensitivity to critical inputs?

While various CMAQ model performance evaluations have been presented elsewhere, the model performance evaluations and sensitivity analyses presented here are most helpful. Since pNO_3 and HNO_3 have such different deposition velocities and are measured separately in CASTNET, it might be informative to show comparisons of the separate modeled species and CASTNET measurements, and perhaps also for the CMAQ and CASTNET estimates of dry deposition of the separate pNO_3 and HNO_3 species, as well as for the CMAQ and CASTNET estimates of TNO3 dry deposition.

c. The utility of the analyses of temporal and spatial variability in the deposition transference ratios (TNOy and TSOx)?

I have a hard time understanding what the spatial variability in these transfer ratios actually means, though it is comforting to see that the patterns seem relatively "smooth" rather than abrupt. Is there is seasonal or diurnal variability in these ratios that might give us a better feeling for what's really going on inside the model (and in the atmosphere)? I wonder if it would be informative to see maps analogous to Figure 5-20 which separately showed the ratios of S conc to S wet dep and S dry dep, and of N conc to N wet dep and N dry dep. Maps showing the ratios of S and N deposition to S and N emissions (perhaps aggregated on a state by state basis) could also be interesting…

The illustrations of the relative absence of temporal variability are comforting, as it is key to have stable regulatory metric which is linearly responsive to emissions changes over time. Some (any) discussion which helped explain the causes and implications (if any) of some of the spatial or temporal variations would be helpful.
17. What are the Panel's views on the areas for future research and data collection outlined in this chapter, on relative priorities for research in these areas, and on any other areas that ought to be identified?

I thought this section of Chapter 7 was especially well done, and well supported by the preceding discussions. A chapter like this should become standard practice in future NAAQS reviews!

18. What are the Panel's views on using an open inlet to capture all particulate size fractions for the purpose of analyzing for sulfate? What is your opinion on using existing CASTNET filter packs as a future Federal reference method for sulfate?

I don't oppose these proposals, although I think the case is somewhat overstated, especially in relation to aquatic acidification effects. A major concern is that this would require exclusive use of CASTNET methods or network and preclude use of fine fraction sulfate measurements which are more abundant, and not demonstrably grossly inferior. Conversely, there's no reason not to include a similar open inlet approach for pNO_3, for which coarse particle deposition may be especially important for N deposition contributions to nutrient enrichment of coastal estuaries. I also think an argument could be made to consider CASTNET HNO_3 and pNO_3 as an (interim) alternative to continuous NOy measurements (more detail on this below and in #9 above).

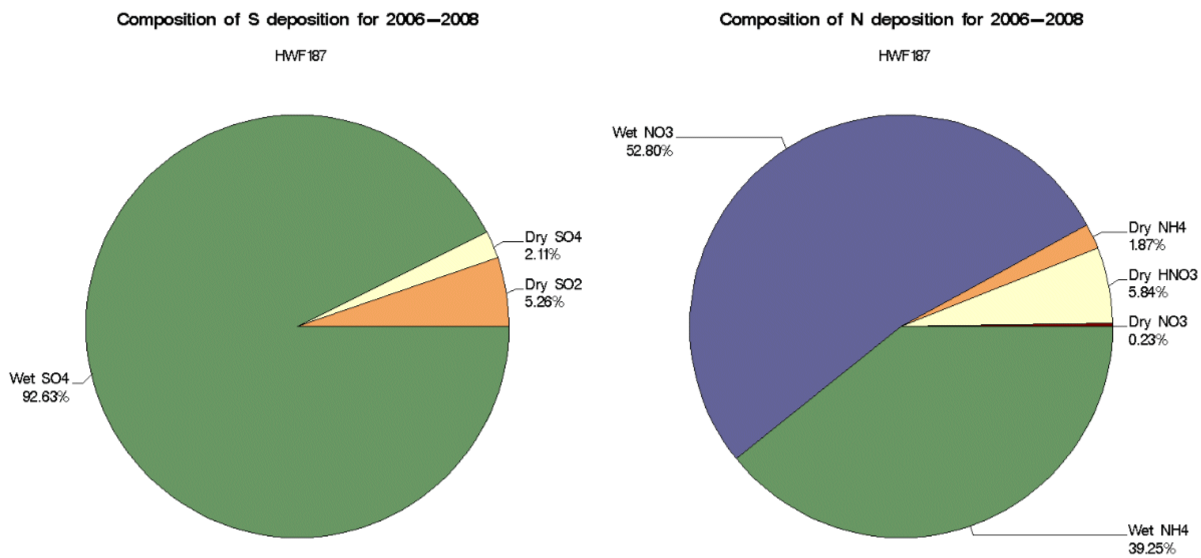
Some counterpoints to the open faced sulfate proposal:

- Away from coastal areas with coarse sea salt or arid or agricultural areas with windblown dust, (and especially in the remote humid, high elevation areas where acidification occurs) there is relatively little coarse sulfate (or coarse nitrate) period.
- The particle cut size characteristics of the open faced collectors have not been well characterized, nor is any information provided on what (small) fraction of the open faced S or N sample is composed of more rapidly depositing coarse mode particles. You need to add a fudge factor, which you could do just as well using fine fraction data.
- Open-faced collectors may take in fog or cloud water. In addition, since coarse particles tend to be alkaline, additional artifacts may occur as gaseous SO_2 or HNO_3 reacts with the alkaline coarse material collected on the sample filter.
- Sulfate and nitrate in coarse mode particles which are formed in the air (and are not sampling artifacts) typically result from reactions of acidic S and N gases and alkaline crustal material or sea salt. Consequently, these particles carry their own cations and represent uniquely well buffered forms of S and N deposition.
- Lastly, the total fractions of S and N depositions from particulate matter – especially in the higher elevations where acidification is an issue – is not very large. Below are the 2006-08 estimates of total S and N deposition for the Huntington Forest CASTNET site (relevant to deposition in the Adirondack case study area). Total

particulate sulfate and nitrate were estimated to account for 2.1% and 0.2% of the total S and N deposition respectively.

As indicated above, I'm not opposed to the proposal to specify an open faced FRM or even the CASTNET filter pack for pSO_4, but think it could also be specified for pNO_3 (assuming problems with loss of volatile NO_3 during summer sampling can be corrected), but also think accommodations could be made (FEM) to accommodate use of fine fraction SO_4 and NO_3 data (with adjustments) to avoid being too prescriptive at this early stage of the NAAQS process, to mandate a compliance network (CASTNET) which is not operated by states but by EPA contractors (funded by $ taken from state monitoring pots), or – in combination with the proposed continuous NOy indicator – to require deployment of a costly new network which may not be currently feasible, or which might indefinitely delay implementation of the NAAQS.

Total Wet and Dry Deposition of S and N at Huntington Wildlife Forest, 2006-2008
Particulate Matter Deposition Accounts for 2.11% S and 0.23% N Respectively



19. What are the Panel's views on requiring measurements of ammonia and ammonium to assist implementation of the standard?

NH_3 and NH_4 measurements would be useful for implementing the sample both directly, to quantify an unregulated but varying element of the compliance metric, and indirectly, to help evaluate and improve emissions inventories and CMAQ model performance. NH_4 measurements are currently available from CASTNET and (urban) CSN networks, and could conceivably be added to IMPROVE. NH_3 measurements are currently very sparse but would be useful – and have added relevance to better understanding sources and trends of $PM_{2.5}$, regional haze, and sources and effects of N deposition on nutrient enrichment. However, I'm not sure current methods have been sufficiently well developed and evaluated for use in routine network operations.

20. What are the Panel's views on having a subset (e.g., 3-5 sites) of monitoring stations in different airsheds that measure for the major NOy species; nitric acid, true NO2, NO, PAN and p-NO3?

Conceptually this is a good idea and can be (and needs to be) justified for reasons beyond just compliance with the proposed NAAQS (for acidification effects). Possibly some of these measurements could be added to existing (or planned) rural NCore sites. NOy, NO and pNO_3 (fine fraction), SO_2 (continuous) and SO_4 (fine fraction) are currently measured at these sites. Add a CASTNET sampler and you've got HNO_3, Open faced (vs fine) SO_4 and NO_3 (so you will know the coarse fractions), and comparative SO_2 by filter pack and continuous analyzer. Adding true NO_2 would be an excellent addition at some sites (I understand there's a photolytic unit currently applying for FEM status), and this would allow calculating NOy minus (NO, NO_2, pNO_3, HNO_3)…

However, while I support the need for these kinds of more detailed measurements at a few sites in a "clustered network" approach, I'm not sure they can be or should be justified just to determine compliance with this secondary NOx/SOx standard or for evaluation and improvement of model performance just for this standard alone. Along similar lines, I'm not sure a large new network of continuous NOy analyzers (at new, remote rural sites) can be justified (or can be afforded, or could be maintained by shrinking numbers of state personnel). I'm uncomfortable with the relatively vague picture of how these new measurements would be conducted. Will this be a state-operated network (like NCore), an enhanced CASTNET network (operated by EPA contractors), an enhanced IMPROVE network – or some combination of the above?

I think a reasonably good argument could be made to specify CASTNET filter pack methods (possibly with some tweaks such as adding NH_3 passive sampler) as the basic monitoring approach, as it does capture the key species – albeit over longer averaging times but which are plenty short enough for the long-term 3-5 year standard. As indicated in the response to question 9 above, I think an alternative T_{NO3*} N deposition transfer function could be developed that would calculate total (CMAQ modeled; or CASTNET + NADP measured) NOx deposition as a function of HNO_3 and pNO_3. If these calculations performed reasonably well, it would allow use of existing and relatively low cost data, use measurements which actually relate to deposition (NOy does not, without a huge assist from the model), and minimize the reliance on complex internal CMAQ calculations. Using this approach, a slightly expanded CASTNET filter pack network might become the initial compliance network, with new CASTNET samplers at a limited number of rural NCore or IMPROVE sites. Continuous NOy could be added to a few of these sites (NCore sites have continuous NOy and SO_2, as well as fine particle SO_4 and NO_3), but should not be required in a large new network. More exotic measurements such as true NO2, PAN, continuous nitric acid, etc should be considered at only a very few "Level 1" type sites.

23. What are the Panel's views on Staff's conclusion that the existing secondary standards for NOx and SOx should be retained to provide protection against direct adverse effects to vegetation due to gas phase exposures?

I agree that existing single-pollutant secondary standards for NO_2 and SO_2 should be retained to protect against direct effects to vegetation due to gas phase exposures.

24. In light of the Panel's views on what constitutes adverse effects to public welfare (see Chapter 3), what are the Panel's views on:

a) the degree to which current levels of NOy and SOx deposition are adverse to public welfare based on evidence and risk information, and information on adversity provided in Chapters 2,3, and 4?

I believe the evidence and risk information provided in previous chapters indicates that environmental damage has occurred and continues to occur as a result of cumulative and continuing SOx and NOx deposition, and that these effects, including acidification of aquatic and terrestrial ecosystems and nutrient enrichment, represent adverse effects on public welfare.

b) target values for ANC that protect against adversity to public welfare in light of the information presented in Chapter 5 concerning levels of ANC and the ecosystem effects associated with those target ANC levels?

ANC is an appropriate environmental indicator of effects from acidification on aquatic ecosystems, and the target levels of ANC being considered - about 50 µeq/L - would represent an appropriate target level that – if attained – could be expected to substantially reduce the adverse welfare effects due to aquatic acidification. A somewhat higher ANC target of say 75 or 100 µeq/L would provide a greater degree of protection for both aquatic and terrestrial ecosystems, although the degree of protection is co-dependent on the target ANC and the target percentage of water bodies (in what sensitivity class or classes) for which the target ANC is to be attained.

c) factors relevant in selecting target percentages of waterbodies to protect at alternative target ANC levels to protect against adverse effects to public welfare, and weights to place on those factors?

Since there can be large variability in the inherent sensitivities of water bodies to acidification effects among different regions and within individual regions, it seems logical to consider protecting a target percentage of lakes from the populations which are potentially susceptible to acidification. The proposed use of Ecoregion III classifications clustered into 5 groups on the basis of ANC seems like an appropriate accommodation of scientific detail without adding unnecessary complexity. As indicated previously, I had a hard time understanding the implications of the different proposed options for selecting target percentages of water bodies for protection. Comparison of the various metrics applied to case study regions where there are sensitive and insensitive lakes which are chemically well characterized would be a useful way to judge the appropriate combinations.

d) alternative standards for NOx and SOx that would protect against adverse effects to public welfare based on the AAPI form, and taking into account

• consideration of target levels of ANC (chapter 5),
• target percentage of water bodies to protect (chapter 5),
• consideration of relevant uncertainties in AAPI components (chapter 7), and
• any other potentially relevant factors, such as levels of co-protection against terrestrial acidification and nutrient enrichment (chapter 6)?

As indicated above, I suggest considering a modification (T_{NOY*}) to the N deposition transfer function such that it would not require extensive new measurements of continuous NOy. I'm not sure it would work well enough, but think it could be considered.

Considering the relatively long time frames associated with acidification and recovery, and also considering that there is no pre-specified time frame for attaining secondary standards, some consideration might be given to standards which require reasonable rates of progress over time toward increasing ANC levels (APPI levels) in water bodies (watersheds) with low ANC levels.

Comments from Dr. Armistead Russell

EPA staff is to be complemented for this work towards developing a Policy Assessment document (PAd or PA) that can be used to support the promulgation of an ecologically-relevant, combined NOx-SOx standard. The PA has evolved considerably since our last review, and the 2nd draft shows that a significant amount of work and additional thought have gone in to its further development. This undertaking demonstrates just how complex it may be to develop multipollutant standards. They are also to be complemented for addressing CASAC's prior comments. They have gotten very far addressing a very complex issue. However, the current document is noticeably not as informative as desired. It is difficult to get the "big picture" of the impacts of choosing different elements of the potential standard. There is little doubt in my mind that this is the most difficult PA (or equivalent document) that I have reviewed.

In the current document, having a clear and comprehensive description of the AAPI is key, as well as the associated components of the AAPI, how the AAPI would be applied, and the consequences of various decisions about the AAPI level, ecoregions and percent of lakes protected, and this makes Chapter 5 a key chapter. At present, however, it does not provide the material needed for someone to read the document and get a clear understanding of all of the concepts, and the tremendous complexities, without a significant amount of work. In part, more data (or distillations of the data) are needed to give the reader an idea of just how big and varied are the various quantities that are being used (e.g., distributions of key variables used to compute the AAPI). Second, sample calculations could be shown. A particular weakness is a demonstration of how the fraction of lakes protected interacts with the choice of ANC. The skeleton is there, but not enough. For example, a more complete demonstration of what went in to making Table 5-12 would be very useful. (Also, as is true in a number of places, some of the variables are not defined or ambiguous, e.g., $DL_{\%ECO}$: is it for NOy+S (I think) or N+S). How do the calculated $DL_{\%ECO}$'s compare with the current estimate of its level? How, specifically, do you find the sites protected from ANC<20 in this case? Further, it would be of interest to see a distribution of $DL_{\%ECO}$ current vs. distributions of $DL_{\%ECO}$ at one of the candidate control levels (e.g., ANC 50, %Prot.=75), along with a description of the decrease in $DL_{\%ECO}$ (again, potentially a frequency distribution). (Figs. A11,16 & 20 are informative, but not as so). It might be even more insightful if a spatial distribution of the control levels were able to be shown (again as % reduction in $DL_{\%ECO}$). It may be necessary to have a set of assumptions for developing that spatial map, and those should be clearly described. It is difficult to see how the administrator and CASAC can provide their best guidance on the level of the standard without further understanding of the level of control that would likely be required to meet various combinations of the level/form/ecosystem choices made to specify the standard. In terms of advising the Administrator, one of the sections that should be strengthened is the one on the approach to defining/choosing ecosystems. While this section is substantial, it was difficult to distill.

I appreciate the chapter on uncertainties. In this analysis, more than those conducted for other standards, the uncertainties in the modeling have to be put forth in a very transparent fashion because those uncertainties impact directly on the translation of the estimated depositional flux to the monitored quantity. They are not small. The magnitude of such uncertainties (e.g., in the transfer ratios) should be quantitative or semi-quantitative. Having an "unknown" for that uncertainty is a weakness of the current document, and indicates an area needing intense assessment. Air quality modeling uncertainty is important to the overall viability of the approach, impacting not only the transfer ratio, but also the estimated NH_3 flux.

The Executive Summary does provide a reasonable overview of the rest of the PA, and is a valuable component of the PA. However, it does suffer from the same elements from which the rest of the PA suffers (e.g., see above for discussions of what is needed to bolster Chapter 5). It also suffers from trying to overly simplify the complexities of the proposed approach. In particular, the AAPI equation really should be included, with explanations of the origin and importance (including magnitude) of each term. Symbols should be defined and figures should have explanatory captions. I would add more headings to show the steps in the conceptual design of an ecologically-relevant standard. I would also note that **at this time** the secondary standard is most strongly supported by the demonstrated relationship between ambient NOx and SOx and aquatic acidification. Like Chapter Five, there is a need to provide more information as to the consequences of making various decisions about the choice of components of the standard.

It is interesting that this PA does not include staff recommendations as to a range of AAPI, % protected or choice of ecosystem approach for the standard (i.e., the elements of the standard upon which we are supposed to give guidance). Similar information has been provided in prior reviews, and that information has been very useful to assess the reasoning behind the choices. As part of our review, it would have been very helpful to see similar information, particularly from people who have been intimately immersed in doing the analyses. This is particularly true given the complexity and novelty of the approach. Further, having only had a rather limited time to review the PA magnifies the problem.

Comments from Dr. David Shaw

This draft demonstrates a marked improvement over the first draft and I feel it is responsive to many of the CASAC member comments. The addition of details with models and uncertainties has resulted in a more informative document.

While this assessment seems to touch on the need for meaningful data from other regions, and it is more specific about what parameters need to be measured to guage the standard, it does not address the resources needed to expand air monitoring into these other regions. I must emphasize the need for data. While this proposed NAAQS is innovative and I appreciate the efforts being made to identify an appropriate NAAQS, it is also model dependent and because of that, it calls for commitments to get better data for future analysis. I am concerned that the USEPA is taking steps towards ranking model data higher than monitored ambient data, and I want to ensure that this is not the direction which NAAQS will take. I believe that real ambient data should be considered in higher regards than model data.

I feel that the PA is a report that should be more readable and user friendly than the highly technical ISA and REA, and in that vein I suggest that it be clear what each indicator represents. For example, the ANC for lakes is relatively easy to measure and therefore represents a large amount of available data, but it doesn't represent streams. In addition, ANC data is typically a summer target which leaves us dependent on models to estimate or adapt for year round use. As a result, I suggest that when ANC is discussed in the broad context, it should be prefaced as lake surface water ANC, not to be confused with stream ANC levels which were not evaluated. On that same note, I suggest that the base cation to aluminum ratio (Bc/Al) be specified as soil water.

Flexibility

There is limited data in regions that currently do not have sufficient monitoring data or modeling efforts to characterize their own sensitive ecosystems at this point. Therefore, I suggest that sufficient flexibility be build into the policy to allow for future monitoring/modeling efforts and characterization.

Models & Data

There are several issues with a heavy reliance modeling. There are differing levels of uncertainties associated with each of the models (thank you for including the updated document on 9/23/2010). One example of these uncertainties is that it is very difficult to assess the dry deposition estimates in CMAQ. As a result, how much confidence do we have in the NO_x/SO_x transference ratios which are based on modeled deposition?

The PA states that the current monitoring networks (IMPROVE, CASTnet, and NADP/NTN) are not adequate to cover all sensitive areas while Chapter 5 suggests that CMAQ will be used to help develop the spatial patterns needed to create the NAAQS. Without sufficient measurements of ambient NO_y and SO_2 in sensitive areas is a serious limitation, again leading to the conclusion that a clear commitment be developed to provide adequate data.

Specific Comments

Brook trout is listed as a sensitive species, it is generally not. I suggest using a more general term like fish which is more accurate. This is also comparable to zooplankton which is not specific either.

ES 12 bullet 1. The statement "at least as protective" does not seem to be appropriate. The secondary standard should is held separate to be protective of human welfare. We most likely will identify different areas of the nation which have different sensitivities. Perhaps something could be added at the end of the sentence to say in the nation, and more protective than the current standard in several parts of the US.

ES- 10 figure caption, delete stream since this model is based on lakes only.

ES 12 bullet 2. Specify lake surface water ANC.

ES 12 bullet 3. Specify soil water Bc/Al values

ES-13 bullet 4. Change to "Less protective against species mortality during acidification episodes"

ES-13 bullet 6. Change may to will. Substitute "fish" for brook trout, because brook trout are not generally considered a sensitive species.

9-18 line 13 change 'may' to 'would'; change 'brook trout' to 'fish'.

A-31-32 Alkalinity section. Text and table need more explanation.

A-34 Line 8. Simplify the explanation, something to the effect that in glaciated areas, the parent material over the bedrock (e.g. glacial till) has been deposited miles from its origin. The soils develop from these parent materials and can be very different from the bedrock.

B-4 Table 1. Suggest identifying the lakes by name, instead of or in addition to their ID number. This would improve the readability and connect the reader to the landscape feature. Likewise with naming the Shenandoah streams (Table 2). Same for Tables 6, 7, 8 & 9.

B-11. Line 29. Change 'lakes" to 'streams'.

Comments from Dr. Kathleen Weathers

Clearly EPA staff has put considerable effort into the conceptual design, analyses, associated uncertainties, and various policy options outlined in this 2nd version of the PA. The stated goal of developing a conceptual design for a standard has been significantly advanced. Many kudos.

It is also true that the timeline was tight for generating this (substantial) revision, and the resultant text is rough and unclear in many places, and lacks well-placed, succinct descriptions of, for example, the AAPI and its component parts (the Introduction notwithstanding; Chapter 5 was a challenge in places).

A few general comments:

That the importance of NHx as a source of pollution/nutrient addition to ecosystems is explicitly considered is an important improvement, even if it must be taken back out of the calculations because of the current regulatory focus on NOx and SOx only.

Perhaps best summarized by the statement on page 5-69: "A critical issue for the Administrator to address in setting a NAAQS based on the AAPI is to consider and weigh the varying degrees of uncertainty in establishing the elements of the AAPI. These uncertainties impact the likelihood that a specific AAPI standard would in fact achieve a target ANC level for a specified percentage of a population of water bodies." While the addition of Chapter 7 is most welcome, I still have some concerns with the large and important influence of CMAQ deposition estimates and as [BC]o, in particular, on the AAPI. Some additional analyses, comparisons, and explanatory text would be helpful in at least bounding these concerns.
For example, some comparisons and analyses that would be likely to either increase or bound confidence in, or understanding of-- CMAQ deposition estimates are still lacking in this document. I do not object to using CMAQ here, in fact, as I said in my comments with the first draft, I think it is appropriate, but since this model is so fundamental to the standard setting, it is critical that the PA is clear about model details, caveats and how it compares to other estimates of deposition (e.g., NADP wet, CASTNET modeled deposition, etc). Despite the fact that CASTNET also models (vs measures) dry deposition, it is instructive to compare the two modeled dry deposition estimates. Similarly CMAQ (or CMAQ/PRISM, as the case may be) vs NADP wet deposition should also be compared. Also, for the case study areas, what are the current deposition estimates and how do they compare to NADP + CASTNET for these locations? I note as well that some of the discussions about the use of CMAQ and its uncertainties often appear defensive, or unnecessarily offensive. And, please, wherever the CMAQ model appears in the text, include the version used.

In regard to the importance of BC (7-30/31) ("largest uncertainty and most influence on the CL"), is there any confidence that can be gained by doing spatial aggregation based on BC? It was dismissed along with bedrock geology by the staff as a way to subdivide or aggregate waterbodies?

In regard to spatially aggregating or dividing waterbodies, The Ecoregions include vegetation and vegetation influences both deposition and the processes involved in Neco, for example. Therefore, I favor them over the one nation approach. That said, 70+ regions seems rather unwieldy.
I think that a few more categories should be added to the monitoring section if a goal is to be able to evaluate the efficacy of the standard. Surface water ANC, and soil monitoring should be included along with field deposition measurements that can extend networks, such as NADP and CASTNET.

A few more specific items:

I'm confused by the first paragraph on page 5-85 in regard to the timeframe consideration. While it is of course important, I'm not exactly sure what, if I were the Administrator, I would do with this statement.

5-53: For specified regions (high elevation watersheds along the Appalachians, for example), occult precipitation can represent a large fraction of N and S deposition, larger than precipitation or dry deposition. I suggest modifying the statement and referencing, for example, MADPro results. Perhaps it is the case that occult precipitation is insignificant at the scale of CMAQ model output, which would be important to note.

7-13: Shenandoah and the Adirondacks are regions of reasonably high heterogeneity with respect to variables that are likely to influence not only the deposition of N and S but all the other parameters in the AAPI index, thus while the statement may be true for large grid cell sizes, it's unlikely to be true over the relevant ecological scales.
9-4 and 9-5—here is an example of a Faulkner-like paragraph whose meaning is completely opaque to me (but I think that there's important information there)

Charge Questions:

Chapter 5 (needs work to make it clearer).

7. What are the Panel's views on the appropriateness of the critical loads that form the basis for the population assessment to determined deposition metrics?

The critical loads concept has been used by EU countries for quite some time; I think it a good and tested framework for considering ecosystem effects of air pollution and around which to build a secondary standard. I find the aggregation method proposed (ES-7) compelling.

a) What are the views of the Panel on the appropriateness of generalizing the f-factor approach to apply to lakes and streams in the Western U.S. and other portions of the Eastern U.S.

The f-factor did not seem well described to me, despite the fact that there were a few places where it showed up in the text. I wished for a "so what" statement on 5-23 with regard to estimation of [BC]o, and what it might mean within the AAPI. So, I'm not sure exactly what the question is here—is the f-factor likely to hold for other locations, or does it matter whether lakes or streams are the target freshwater (given the divergences in Fig. 5-6b)? As noted above, the [BC] is a critically important variable.

b) What are the views of the Panel on the filtering criteria used to remove lakes and streams that are naturally acidic or not sensitive to atmospheric deposition?

These are important and appropriate criteria on which to filter freshwater systems from the database. This filtering is responsive to the Panel's previous recommendation.

8. What are the Panel's views on the suggested methods for determining appropriate values of reduced nitrogen deposition in establishing NOx/SOx tradeoff curves?

As with all of the deposition estimates, CMAQ is the source, and there aren't really any other great options. NADP chemistry and PRISM precipitation might be used to get at wet inputs (or compare them with CMAQ). The inclusion of NHx is important.

CHAPTER 9

23. What are the Panel's views on Staff's conclusion that the existing secondary standards for NOx and SOx should be retained to provide protection against direct adverse effects to vegetation due to gas phase exposures?

That there are adverse effects to vegetation as a result of exposures to SOx and NOx has been established and is reiterated in this document. However, isn't it likely that the proposed new standard would result in concentrations at, or lower than the current concentrations, and therefore be protective? Or, is the question about whether these existing secondary standards should remain in place in addition to a deposition standard? (I do think that protection against adverse effects to vegetation due to gas phase exposures should continue.)

24. In light of the Panel's views on what constitutes adverse effects to public welfare (see Chapter 3), what are the Panel's views on:

a) the degree to which current levels of NOy and SOx deposition are adverse to public welfare based on evidence and risk information, and information on adversity provided in Chapters 2,3, and 4?

Research has shown that acidifying atmospheric deposition at its current levels results in ecosystem functional changes, and aquatic acidification, in sensitive ecosystems; the information in Chapters 2-4 reinforces this.

b) target values for ANC that protect against adversity to public welfare in light of the information presented in Chapter 5 concerning levels of ANC and the ecosystem effects associated with those target ANC levels? These levels are supported in the literature; they are defensible and appropriate.

c) factors relevant in selecting target percentages of waterbodies to protect at alternative target ANC levels to protect against adverse effects to public welfare, and weights to place on those factors?

It seems to me that the challenges of creating and enforcing a spatially explicit standard at fine spatial scales will be large. The graphical representation of percentages of waterbodies that would, or would not be protected is very informative and suggests that an aim of 50% is too low. But this is clearly a value judgment. What are the socio-ecological characteristics of the waterbodies that would not be protected? Are they public water supplies? High elevation waterbodies?

d) alternative standards for NOx and SOx that would protect against adverse effects to public welfare based on the AAPI form, and taking into account • consideration of target levels of ANC (chapter 5),

These appear to be consistent with what's in the literature (see especially Driscoll et al. 2001), and what others have used for policy targets.

• target percentage of water bodies to protect (chapter 5),
The ranges presented are wide; I cannot come up with rationale to suggest different targets, however.

• consideration of relevant uncertainties in AAPI components (chapter 7), and

Thank you for chapter 7. As noted, almost all AAPI components have significant uncertainties, Transference and BC, especially. I continue to wonder whether uncertainties are exacerbated, or could be reduced, by choosing appropriate spatial scales for aggregation (see comments from last review). Also, I still find not wholly adequate the discussion and analysis of the CMAQ model (see above). While I agree that it is an appropriate model to be used for examining deposition across the US, the uncertainties that have to do with estimating deposition to heterogeneous terrain were not addressed, for example.

The elasticity analysis was an interesting way to examine uncertainties. It was not clear to me why 1% was chosen for each of the variables, however.

• any other potentially relevant factors, such as levels of co-protection against terrestrial acidification and nutrient enrichment (chapter 6)?

I agree that using ANC as a target for chemical protection is a good one, and a defensible place to focus. Further, by definition and as pointed out, there will be some level of protection afforded to the adjacent terrestrial systems that influence downstream freshwater systems. The analysis of protection that would be afforded for terrestrial systems using Bc:Al was very useful. However, since data are limited, and the linked biogeochemical reactions within terrestrial ecosystems as a result of N and S deposition are complex, and may have opposite effects (nutrient enrichment vs acidification), I do not think that addressing co-protection further than what has been accomplished in this document is warranted. It will be crucial, however, to monitor terrestrial ecosystem responses to changing deposition scenarios so that sufficient data are available for the next review.