



UNITED STATES ENVIRONMENTAL PROTECTION AGENCY  
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June 22, 2010

EPA-CASAC-10-014

The Honorable Lisa P. Jackson  
Administrator  
U.S. Environmental Protection Agency  
1200 Pennsylvania Avenue, N.W.  
Washington, D.C. 20460

Subject: Review of the *Policy Assessment for the Review of the Secondary National Ambient Air Quality Standards for NO<sub>x</sub> and SO<sub>x</sub>: First Draft (March 2010)*

Dear Administrator Jackson:

The Clean Air Scientific Advisory Committee (CASAC or Committee) NO<sub>x</sub>-SO<sub>x</sub> Secondary NAAQS Review Panel met on April 1-2, 2010, to review EPA's *Policy Assessment for the Review of the Secondary National Ambient Air Quality Standards for NO<sub>x</sub> and SO<sub>x</sub>: First Draft*. The chartered CASAC held a public teleconference on May 3, 2010, to review and approve the report. This letter provides CASAC's overall comments and evaluation. We highlight the most important issues which need to be addressed as the first draft Policy Assessment (PA) is revised. The CASAC and Panel membership is listed in Enclosure A. The consensus responses to EPA's charge questions are presented in Enclosure B. Finally, Enclosure C is a compilation of individual panel member comments.

Overall, the CASAC NO<sub>x</sub>-SO<sub>x</sub> Secondary Panel found that the first draft of the PA has begun to provide a framework for developing a secondary standard that integrates two criteria pollutants in a valid, scientifically based approach and that also accounts for the presence of chemically reduced forms of nitrogen (NH<sub>x</sub>). EPA staff continues to innovate in the face of many complexities. We realize that this complexity has led to some portions of the PA that are incomplete and others that need further refinement, and we look forward to reviewing subsequent and more complete drafts. The Panel is supportive of the approach that EPA staff has followed in their development of an Atmospheric Acidification Potential Index (AAPI), which integrates the effects of NO<sub>x</sub> and SO<sub>x</sub> deposition on aquatic acidification. This index can be the basis of a standard that protects sensitive ecosystems while allowing for the actuality that in most locations in the U.S. NO<sub>x</sub> and SO<sub>x</sub> deposition may not be causing substantial harm.

In addition to finishing the sections of the PA that EPA has identified as being incomplete, a specific need identified by CASAC is more comprehensive sensitivity and uncertainty analyses of the proposed approach, and we recommend that a separate chapter be devoted to such analyses. We recognize that a probabilistic uncertainty analysis is likely not practical given the time constraints to complete the PA and development of a secondary standard. The reasoning that supports EPA's assessment of the relative values of the uncertainties should be provided. At present, the reasoning behind the rankings of the uncertainties are not clearly articulated. Sensitivity analyses of the various terms included in the AAPI equation should be performed. The sensitivity analyses should be guided by estimates of the potential range of the terms in the AAPI. For example, the two approaches for

calculating  $N_{eco}$  can be used, and results from multiple CMAQ (and other air quality models and data analysis efforts) can be used to define potential ranges for  $L(NH_3)$ ,  $Q$ ,  $VNO_y$  and  $VNO_x$ . The Panel recommends the “V” terms be changed for clarity. The Panel also recommends that the “g(.)” term of the AAPI equation be expanded to show the nitrogen cycling and background cation weathering terms explicitly.

The approach laid out by EPA staff relies heavily on results of Community Multiscale Air Quality (CMAQ) modeling. Given this dependence, the PA should provide a more thorough evaluation of the ability of CMAQ to simulate the ambient concentrations and wet and dry deposition fluxes of the  $SO_x$ ,  $NO_x$  and  $NH_x$  species used in determining the AAPI. While details of this evaluation can be contained in an appendix, summary results should be brought into Chapter 4. Additionally, the separate, recommended chapter on uncertainty should use this information in the overall assessment of uncertainty.

The Panel acknowledges that the current focus of the PA is on adverse impacts of  $NO_x$  and  $SO_x$  deposition in acid sensitive ecosystems. However, the PA should also include a succinct discussion of other potential impacts of  $NO_x$  deposition to N-limited ecosystems. For example, increased N deposition to N-limited ecosystems can lead to production increases that may be either beneficial or adverse depending on the system and management goals.

In revising the PA, the Panel asks that EPA use a consistent set of units in its presentation of data, results and equations. Given the focus of the document on depositional effects that depend largely on the number of moles of reactive N or oxidized S, use of SI units (International System of Units) is likely the best approach. It may be most facile in some cases to just provide the necessary arithmetic conversion necessary to produce the results in SI units to avoid the problems associated with extensively redoing tables and figures. The units in every equation variable should be stated.

In closing, the Panel was pleased with the development of the PA and trusts that our comments are useful in the PA’s revision. We look forward to seeing the revised PA, along with the Agency’s response to how our comments have been addressed.

Sincerely,

*/Signed/*

Dr. Armistead (Ted) Russell, Chair  
CASAC  $NO_x$ - $SO_x$  Secondary Panel

*/Signed/*

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

Enclosures

## **NOTICE**

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## Enclosure A

### Rosters of the CASC NO<sub>x</sub>-SO<sub>x</sub> Secondary Review Panel and CASAC

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

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## Enclosure B

### CASAC's Consensus Responses to EPA's Charge Questions

#### Executive Summary

The Panel finds that the draft Executive Summary is well written and is an appropriate representation of the main Policy Assessment document. For the most part, the technical language, figures and the AAPI formulae convey the essence of each chapter. Clearly, future versions would need to incorporate the changes in the evolving main policy document. The fourteen-page Executive Summary is somewhat long. The summary of Chapter 5, the Conceptual Design of an Ecological Standard, should be improved to effectively communicate the significant new concepts such as SO<sub>x</sub>-NO<sub>y</sub> deposition tradeoff and deposition load function. In particular, the equation for the proposed standard (AAPI) needs to be explained more clearly, maybe with the aid of a conceptual diagram specifically representing the underlying rationale of each of the terms in the proposed AAPI equation.

#### Chapter 2: Known or Anticipated Ecological Effects

*1. What are the views of the Panel on the appropriateness of staff's weight-of-evidence approach which assesses information from across the various ecological research areas described in the NO<sub>x</sub>/SO<sub>x</sub> Secondary Standards Integrated Science Assessment (ISA), including studies of acidification and nutrient-enrichment in aquatic and terrestrial ecosystems, and laboratory research on responses of plant and animal species to acidification and nutrient enrichment? To what extent is the presentation of evidence drawn from the ecological effects studies assessed in the ISA technically sound, appropriately focused and balanced, and clearly communicated?*

The current document provides a good review based upon the ISA. There needs to be clarification, however, with respect to some of the details including how the document provides results with respect to the actual years of the analysis. Some of the discussion is repetitive and additional editing is needed to decrease the redundancy. The Panel asks that the referencing to the scientific literature and prior review documents be consistent throughout the PA. Statements of fact need to be referenced to the literature.

The text is often written to suggest that acidification or nutrient enrichment have the potential to affect all ecosystems in a similar manner. The document needs to clarify that many effects being discussed are applicable only to the most sensitive components of ecosystems. These ecosystems may warrant special protection, but they do not represent a response that is universal for all ecosystems and their respective components.

*2. To what extent are the interpretation and presentation of the results of the exposure and risk assessment technically sound, appropriately balanced, and clearly communicated?*

The staff has adequately summarized the information contained in the REA, but some further clarification of the major points is warranted. Most of the assessments presented use steady state

models; ecosystems, however, are never in steady state. A summary of the assumptions and limitations of the steady-state models used in the ecological effects studies should be included in the document.

*3. What are the views of the Panel on the critical uncertainties associated with the risk and exposure analysis and the evidence from ecological effects studies that need to be characterized in terms of their potential implications for the secondary standards?*

The discussion on critical uncertainties (Pages 35-37) requires a more rigorous treatment than that included in the current draft. The rankings of uncertainty (high, fairly high, intermediate or low confidence) are based on EPA Staff judgment without any explanation of how the Staff arrived at those rankings. For example, have the models and input parameters been adequately evaluated to assign high confidence in them? If so, it is essential to provide justification for the assigned rankings. In addition, it is recommended that sensitivity analyses be conducted for the different models used for aquatic acidification and terrestrial acidification and presented in the document (please note our earlier comment on the need for a separate chapter on uncertainty).

### **Chapter 3: Considerations of Adversity to Public Welfare**

*4. What are the views of the panel regarding the characterization of adversity to public welfare presented in this document? What are the views of the panel regarding the use of the ecosystem service framework as an additional metric to inform questions of adversity? What are the views of the panel regarding the usefulness of including economic valuation of some of these ecosystem services in the policy assessment document?*

In Chapter 3, EPA needs to describe for the Administrator and the public how and why the current effects of SO<sub>x</sub>/NO<sub>y</sub> deposition on sensitive ecosystems matter in terms of services and functions of ecosystems that are important to people. This is the motivation for this review. The ecosystem services framework and the available economic valuation results can help, but the Chapter does not yet provide sufficient information on the ecosystem services and the resultant impacts of SO<sub>x</sub>/NO<sub>y</sub> deposition. Throughout the chapter it is important to re-iterate that ecosystem effects of concern as a result of SO<sub>x</sub>/NO<sub>y</sub> deposition are currently occurring only in certain sensitive areas.

The economic valuation estimates are useful when they are tied to the change or loss in value of ecosystem services that can be reasonably attributed to SO<sub>x</sub>/NO<sub>y</sub> deposition. It is true that neither the economics nor the science can define precisely the exact threshold at which an effect becomes adverse, but the relative values of available estimates still provide useful information to assess the question of adversity when they are put in the proper context. The monetary values that are presented in the chapter need more explanation as to their policy implications. A table summarizing the monetary valuation associated with various ecosystem services estimates would be helpful.

*5. To what extent is the presentation of ecosystem services in this document scientifically sound and clearly communicated?*

The basic concepts of ecosystem services and how they relate to ecosystem function and to the idea of welfare effects as defined in the Clean Air Act are clearly communicated and well presented. The description and summary of how these services are specifically being affected by SO<sub>x</sub>/NO<sub>x</sub> deposition need to be improved. The basic concepts of ecosystems services are useful to explain why the health of these systems matters, but more discussion is needed beyond providing the conceptual framework. This framework needs to be used to describe the effects for which the science provides reasonable confidence on the deleterious impacts that are affecting sensitive ecosystems under current conditions, and not just describing the effects that could happen. The description of deleterious ecosystem effects needs to be placed in the context of their geographical distribution. More can be done to characterize the importance of the current observed effects even though most changes in ecosystem services cannot be completely quantified.

The presentation lacks a clear communication of the potentially beneficial effects of nitrogen inputs to nutrient limited ecosystems. Due to their inherent capacities for N retention, the majority of US ecosystems will not experience significant negative impacts from N-induced acidification or nutrient enrichment. Such a reality doesn't eliminate the benefits of protecting more sensitive and valuable but less common ecosystems, but EPA will be open to criticism if this is not explicitly acknowledged. Finally, timber products should be added to Table 3-1.

*6. What are the views of the panel on the critical uncertainties associated with articulating adversity to public welfare that need to be characterized in terms of their potential implications for the secondary standards?*

It is quite clear that current levels of acidification are causing adverse effects on aquatic resources in some locations. It seems there is sufficient evidence to support that these effects are adverse to public welfare with respect to a substantial proportion of lakes and streams in sensitive regions. These regions cannot support several species of fish and other aquatic biota due to anthropogenic induced acidification. Key uncertainties include how much of the adversity would be reduced with a standard based on a given level of acid neutralizing capacity (ANC). The economic valuation estimates show additional value for incremental improvements in ANC, but do not reflect all the benefits of decreased deposition or point to a specific ANC level that should be required. Related to this is the question of what percentage of acidified lakes and streams need to achieve these ANC levels to eliminate or at least reduce the adverse effect to an acceptable level. The evidence may not provide definitive answers to these questions, but the implications of available evidence can be more explicitly stated with regard to these questions and will be important considerations to inform the Administrator's policy decisions on this NAAQS.

## **Chapter 4: Addressing the Adequacy of the Current Standards**

*7. What are the views of the Panel on staff's assessment of the adequacy of the form of the existing NO<sub>x</sub> and SO<sub>x</sub> secondary standards? To what extent does the Panel agree with staff's assessment of the protection provided by existing standards, given the current levels, forms, averaging times, and indicators?*

In Chapter 4 clear and convincing arguments are presented questioning the ecological relevance of the current SO<sub>2</sub> and NO<sub>2</sub> secondary standards (and many of their individual components), and concludes that current standards do not adequately protect against adverse environmental effects from SO<sub>x</sub> and NO<sub>x</sub>. Well-documented observations of chemical and ecological effects of S + N acidification in monitoring programs like TIME, LTM and EMEP, combined with the observations of no exceedances of current NO<sub>2</sub> or SO<sub>2</sub> standards in areas experiencing effects of aquatic or terrestrial acidification or nutrient enrichment provide a direct indication that current standards are not adequate. As the environmental effects from SO<sub>x</sub> and NO<sub>x</sub> occur primarily through the long-term cumulative deposition of multiple S and N compounds, combined together rather than individually, the Chapter persuasively shows how the components of the current secondary standards are inappropriate in terms of indicators, averaging times, levels and forms, as well as in their single pollutant approaches to multi-pollutant problems.

*8. What are the views of the Panel on the time frame of ecological response related to current deposition? The adequacy evaluation relies on recent NO<sub>x</sub>, SO<sub>x</sub>, deposition, and on long-term steady state ANC. Does the panel agree that long-term steady state ANC is the most appropriate representation of ANC for evaluating the adequacy of the current standards?*

Use of long-term ANC derived from steady state modeling and recent deposition rates of SO<sub>x</sub> and NO<sub>x</sub> (and NH<sub>x</sub>) to evaluate the adequacy of current standards is a reasonable approach. This approach could be enhanced by also applying dynamic acidification modeling techniques to “hindcast” changes for comparison with recent measurement or paleo-limnological data, as well as to forecast chemical and ecological responses if current deposition rates remain unchanged. A parallel dynamic modeling effect would greatly inform the appropriateness of the assumptions invoked in steady-state calculations. Current levels of acidification and associated effects result from a combination of current and historical deposition of SO<sub>x</sub> and NO<sub>x</sub> compounds, and their cumulative effects on the chemical and biological environment. As current deposition rates are lower in most locations than they were several decades ago, it's likely that some aquatic and/or terrestrial ecosystems have shown some signs of recovery. An assessment of the adequacy of current standards should consider the extent to which and rates at which some less sensitive systems may continue to recover at current S + N deposition, while other more sensitive systems will not recover or may experience further deterioration if current “non-sustainable” rates of deposition remain unchanged.

9. *To what extent are the characterizations of ambient air quality and deposition appropriately characterized, relevant to the review of the secondary NO<sub>x</sub> and SO<sub>x</sub> NAAQS, and clearly communicated?*

The characterizations of air quality and deposition presented in Chapter 4 are, for the most part, appropriate and relevant to the review of secondary NO<sub>x</sub> and SO<sub>x</sub> standards. While the Chapter is nominally focused on evaluating the adequacy of current standards, it also includes considerations of the potential to use current monitoring networks (supplemented by additional measurements and/or by CMAQ model results) for the development of potential new standards and/or determining compliance with them. While most of the geographical distribution information included in maps is useful and clearly presented, several of the figures appear to have inaccurate legends or captions (Figures 4-8 through 4-11). It would also be useful to include additional figures showing the geographical distribution of total NH<sub>x</sub> N deposition, as well as the fraction of total N deposition contributed by chemically reduced (or oxidized) N.

There are some important figures showing the relationships between atmospheric concentrations and total deposition of S and N ( $V_{S/N}$ ) that are not well developed in Chapter 5 (e.g., Figure 5-5). These types of depictions and the relationships they demonstrate are important. The importance of these relationships needs to be emphasized and their importance expanded upon in Chapter 4. Presumably, it could be clearly shown, through use of CMAQ ratio maps and scatter plots based on grid point comparisons, that NO<sub>y</sub> is a much better predictor or indicator of total oxidized N (NO<sub>x</sub>) deposition than is NO<sub>2</sub>, and hence this result would directly support the evaluation of adequacy of the various approaches for estimating S and N deposition inputs. This chapter should also include a summary of the performance assessment of CMAQ, as discussed in response to charge question 18.

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

10. *What are the views of the Panel on staff's proposed conceptual framework for the structure of a multi pollutant, ecologically relevant standard for NO<sub>x</sub> and SO<sub>x</sub>? 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 NO<sub>x</sub> and SO<sub>x</sub>?*

The proposed conceptual framework for the structure of the proposed standard is very well-thought out and quite innovative in its design for addressing complex linkages between various components (ecological effects, aquatic chemistry, atmospheric wet and dry deposition, and atmospheric concentrations of NO<sub>y</sub> and SO<sub>x</sub>). However, as currently proposed, calculation of AAPI assumes deposition of reduced forms of reactive nitrogen to be constant in the future which is unlikely to be true considering the upward trajectory of U.S. ammonia emissions. A more realistic approach would be to treat ammonia as a variable component of the AAPI standard that can be updated. Additionally, the underlying assumption of steady state behavior should be evaluated with a parallel effort using sensitivity analysis and dynamic modeling. The chapter should be improved to effectively communicate the important new concepts such as SO<sub>x</sub>-NO<sub>y</sub> deposition tradeoff and deposition load function. The AAPI equation for the proposed standard should include the geographical extent and numerical range of its applicability.

*11. What are the views of the Panel on the relevance of the conceptual design for developing standards to protect against aquatic acidification effects?*

*a) What are the views of the Panel on staff's proposed options for the ecosystem acidification model to represent the ecological response function?*

The Panel agrees with EPA staff that steady-state models are probably the best approach to develop a combined NO<sub>x</sub> and SO<sub>x</sub> secondary standard to protect surface waters against the effects of acidic deposition at this time. The Panel recommends, however, that the EPA take advantage of on-going efforts in dynamic modeling. Dynamic model calculations coupled with long-term measurements would greatly inform the regional and national steady state modeling effort by helping to evaluate the assumptions invoked in the application of steady-state models, in evaluating the relative sensitivity of aquatic and forest ecosystems from acidification by acidic deposition, and building experience in the use of dynamic models in critical load calculations for future assessments.

*b) What are the views of the Panel on the relative merits of the two techniques for calculating Neco, the parameter representing the amount of deposited nitrogen that is available for acidification due to uptake, denitrification, and immobilization.*

In principal, Equation 3 (page 159) captures many of the major landscape and ecological factors that influence the processing of atmospherically deposited N within an ecosystem, including biological and abiotic retention of N (immobilization, uptake and sedimentation), and gaseous loss (denitrification) after N has been deposited and/or transported. This approach opens the “black box” and attempts to estimate some of the component parts of N processing and loss. It is best modeled as a dynamic process because such factors as age and stage of vegetation, soil moisture, moisture regime, and nutrient demand will affect various components of N cycling. For many ecosystems, complete supporting data may not be available.

Equation 4 is basically a mass balance approach that keeps track of inputs and outputs. It does not include the detailed biogeochemical processes identified in Equation 3. Data for this latter approach may be much more readily available than the approach using Equation 3. Watershed N retention can be estimated using this approach (e.g., Lovett et al. 2000, and many others), but the processes involved in the retention are not detailed. This approach is most effectively used when hydrologic boundaries (e.g., watershed) are defined. Hence, the Panel recommends that the mass balance (i.e., Equation 4) approach should be used.

*c) what are the views of the panel on staff's proposed options for developing acid-sensitivity classes to categorize the national landscape? Is it appropriate to base this classification on bedrock geology? Should multiple criteria be used to inform the sensitivity categories.*

Maps of bedrock geology are likely to be most effective in indicating the sensitivity of acid-sensitive ecosystems in unglaciated parts of the country. Where glacial till or other forms of surficial materials have a major influence on soil characteristics, the direction contribution of the

bedrock geology may be decreased. Ideally, a combination of topographic and landscape criteria could be used that includes such factors as elevation and landuse/landcover as well as bedrock and surficial geology. For example, highly N-deficient, young forests are far more likely to take up large amounts of deposited N than older forests and/or those nearing N saturation (the latter being mostly at higher elevations). In the case of S, deficiencies are very unlikely and thus soil chemical adsorption is the most likely cause of retention of atmospheric inputs (if it occurs). In any event, sensitivity to acidification is a function not only of the ability of the soils to buffer incoming acids but also the ability of both soils and living biota to retain incoming S and N.

*d) What are the views of the Panel on staff's proposed options for how to aggregate acidification modeling from the catchment-scale to represent acid-sensitivity categories at the national-scale?*

It is difficult to evaluate these options in the abstract, without seeing comparative example results from applying alternative approaches. The approach of specifying a percentage of water bodies to be protected within a given area appears reasonable, but would be critically dependent on how the areas were selected. Selecting a specific percent of lakes or stream segments to protect seems somewhat arbitrary, especially when combined with the range of approaches for selecting the size, variability and distribution of lake and stream characteristics within a region. It might be useful to consider approaches by which the water bodies within any specified area could be first stratified to include only those which were considered potentially susceptible to acidification, based on estimates of preindustrial ANC, or acid sensitivity class. Then a decision to protect a specific fraction of the potentially susceptible surface waters within that area would be more objective.

*e) What are the views of the Panel on staff's suggested method to account for reduced nitrogen in the deposition metric?*

As we note in response to Charge Question 10, even though the framework and the structure of proposed approach "takes into account" the reduced ambient nitrogen and its deposition in designing AAPI (atmospheric acidification potential index), it does so by including current ammonia/um levels. This approach would lead to over or under-prediction for an area depending upon whether the actual concentrations of NH<sub>x</sub> increase or decrease over time (see also response to charge question 10).

*12. What are the views of the Panel on the relevance of the conceptual design for developing standards to protect against terrestrial acidification effects? terrestrial nutrient enrichment effects? aquatic nutrient enrichment effects? Does the Panel have suggestions on additional data or methods that might enable EPA to expand the current aquatic acidification approach to cover additional effects?*

Useful standards for protecting against adverse acidification or nutrient enrichment response need to be tailored for the biological and physicochemical characteristics of sensitive ecosystems. Recognition that a more complete data set is available to support the development of an aquatic effects standard, and that such a standard would likely demand lower levels of

nitrogen and sulfur inputs to ecosystem, EPA staff is advised to focus on an AAPI standard driven by aquatic effects concerns.

The proposed conceptual design approach for terrestrial acidification effects is along the lines of the approach for aquatic acidification and seems reasonable and consistent with state-of-the-science. The “BC/Al” ratio appears to have the same level of robustness as a terrestrial ecological indicator (directly measurable and predictable via models) as ANC has for lakes and streams. It meets the important criterion that it is directly responsive to deposition changes as well as being a good and direct predictor of ecological effects. The range currently considered (0.6 to 10 B<sub>C</sub>/Al) seems rather broad compared to ANC range of 50 to 100 µeq/L and, ideally, might be narrowed in future assessments. If appropriate, the indicator for the more sensitive ecosystem could be used to provide protection for the less sensitive ecosystem. However, this would require further evaluation.

For nitrogen nutrient enrichment of both aquatic and terrestrial ecosystems, we recognize it will take additional effort before one can establish appropriate ecological indicators for aquatic and terrestrial systems and associated secondary standards. One approach worth investigating for its applicability to the current effort is the important effort that is currently taking place in EPA’s total maximum daily load (TMDL) process of deriving and apportioning pollutant loading limits for impaired surface waters. EPA should look into combining the TMDLs and secondary NAAQS for both acidification and nutrient enrichment effects.

For the case of nutrient enrichment of coastal estuaries, it would be useful to increase the level of confidence (through additional modeling and measurements) in the estimates of the fraction of reactive N (including reduced nitrogen, NH<sub>x</sub>) loading that is attributable to atmospheric deposition.

*13. What are the views of the Panel on the critical uncertainties associated with the conceptual design of an ecologically relevant multi-pollutant standard that need to be characterized in terms of their potential implications for the secondary standards?*

This is an important question and it correctly focuses on the implications of uncertainties for setting the secondary standards rather than the uncertainties by themselves. However, the discussion of “uncertainties” in this draft (in this chapter and throughout the report) is generally descriptive in nature, which is not helpful in providing a quantitative effect of uncertainties on the potential effectiveness of the secondary standards. It will be helpful if all the uncertainties and their implications are described in one place under a separate chapter (please note our earlier comments above). It is recommended that EPA conduct a sensitivity study to characterize uncertainty associated with different components of the conceptual framework and propagate that uncertainty at every step to arrive at an ensemble of SO<sub>x</sub> and NO<sub>y</sub> response surfaces to meet a given standard. This would in turn provide a range for SO<sub>x</sub> and NO<sub>x</sub> concentrations that might satisfy a given standard.

As an example, the calculations for Adirondacks could be repeated by using the following approach:

- Calculate Neco using different proposed approaches,
- Use a probable range of estimates for  $[BC]_0$ ,
- Use an estimate of Q based on different years (dry vs. wet year),
- Calculate  $L(NH_x)$ ,  $V_{NOy}$  and  $V_{SOx}$  using different air quality model simulations that may already be available,
  - Use CMAQ, CAMx, or other model simulations for different years (with different meteorological conditions and different emissions),
  - Use different chemical mechanisms, if available.

*14. To what extent do the figures and examples aid in clarifying the text? Should more or less information of this type be included in the second draft?*

The Panel notes that figures, tables, maps, and examples were very useful in understanding the text material, especially when subjects discussed are complex. If anything, we would have preferred more tables, figures, and examples as appropriate. In particular, a separate conceptual diagram should be added which shows the important contributing factors for each term in the AAPI equation.

## **Chapter 6: Options for Elements of a Standard to Protect Against Effects from Aquatic Acidification**

*15. To what extent does the Panel agree that the proposal to develop an Atmospheric Acidification Potential Index (AAPI) linked to key determinants of aquatic ANC is a reasonable approach to developing an ecologically relevant standard? Does the Panel generally agree that the secondary standard options identified by staff (including indicator, averaging time, form, and level) are generally consistent with the available scientific and technical information and are appropriate for consideration by the Administrator?*

The CASAC Panel supports the Atmospheric Acidification Potential Index (AAPI) as a reasonable approach to developing a  $NO_x$  and  $SO_x$  Secondary Standard to protect against aquatic (and potentially terrestrial) acidification. This or something similar to this steady-state approach is the only practical method that could be used for a national assessment within the current timeframe of policy recommendations and development. We agree that the indicators ( $SO_x$  concentrations,  $NO_y$  concentrations, ANC) selected are appropriate. A multi-year averaging time should be considered given the year-to year climatic influence on atmospheric deposition. The Panel agrees that the multi-pollutant approach is appropriate for the  $NO_x$  and  $SO_x$  secondary standard to protect ecosystems from acidic deposition. It would be useful to conduct a parallel effort to apply/use dynamic models and surface water time-series data to evaluate the limitations in steady-state approaches and assumptions in developing the acidification secondary standard.

*16. What are the views of the Panel on the degree of uncertainty associated with each element of the suggested standard, e.g. the ecological indicator; the concentration to deposition ratios, reduced nitrogen, the natural background ANC, and the ambient indicator and averaging time for NO<sub>x</sub> and SO<sub>x</sub>, and its relationship to the degree of protection that could be expected from the standard? What are the views of the Panel on how to fairly characterize the uncertainty associated with the degree of protection that such a standard would provide from aquatic acidification?*

The Panel clearly recognizes the challenges in the development of the AAPI. There are considerable uncertainties associated with all steps of developing an AAPI and the implementation of the various modeling processes involved in each step of the analysis, from ecological effects to deposition to ambient NO<sub>x</sub>/SO<sub>x</sub> concentrations. Information on levels of ANC protective to fish and other aquatic biota has been well developed and presents probably the lowest level of uncertainty in the entire methodology. However, there is still high uncertainty regarding pre-industrial levels of ANC. In addition, estimates of ANC could be flawed by the application of steady-state models that assume that the soil exchangeable base cations remain consistent over time. In reality, the base saturation of soil decreases with acidic deposition, natural leaching processes, and plant base cation uptake and therefore the use of dynamic models should be considered to reduce this source of uncertainty. The modeled concentration to deposition ratios (or “atmospheric deposition transformation ratios”) may present a high level of uncertainty, which should be evaluated in more detail with comparison to observations. Various individual components behind the total V<sub>N</sub> or V<sub>S</sub> are not very well defined and could be quite variable especially in complex mountain terrain and non-uniform canopies. Deposition velocities of reactive N species, such as NH<sub>3</sub> and organic N, both for the aquatic and terrestrial ecosystems, have high levels of uncertainty. Information on ambient levels and distribution of the reduced N forms, NH<sub>3</sub> and NH<sub>4</sub> are scarce which increases the uncertainty of the modeled ambient concentrations and deposition of these pollutants, especially in remote areas. Strong reliance on using the CMAQ model would be greatly improved by more specific comparisons between CMAQ estimates and measured values. This is especially true for the modeled values of NH<sub>3</sub> levels and distribution. Therefore more emphasis should be placed on improvement of the model performance evaluation. In that regard recent efforts of EPA and National Park Service (NPS) CASTNET network on adding NH<sub>3</sub> measurements with passive samplers are applauded and should be supported.

The Panel recommends that EPA pay close attention to an issue of matching temporal and spatial scales in various components of the AAPI.

Generally, a higher level of uncertainty is expected for the terrestrial ecosystems versus aquatic ecosystems due to their more complex structure and diversity throughout the US, especially for the nutritional effects of deposition of the total reactive N transformed into the NO<sub>x</sub> concentration-based standards. Because uncertainties associated with various elements of the standard have not been clearly delineated, it is difficult to state with any degree of confidence what degree of protection could be expected from a given standard. One possible approach is to characterize uncertainty by conducting a sensitivity analysis for each chemical form of the standard and propagate these uncertainties to arrive at a range of protection that a given standard would provide from aquatic acidification.

*17. What are the views of the Panel on aggregating the terms ( $Q$ ,  $N_{eco}$  and  $BC_0^*$ ) used in estimating natural background ANC, denoted by the function  $g(\cdot)$ , into 5 bins based on the geologic classification scheme?*

The Panel believes strongly that there are few advantages associated with aggregating the  $g$  term in the AAPI equation. The Panel suggests that  $BC_0^*$  and  $N_{eco}$  be separated and addressed separately. This separation makes the role and uncertainty of these processes in regulating the AAPI equation more transparent. The Panel also suggests that the EPA consider and possibly include a term within  $g$  which accounts the supply of naturally occurring organic acids to the natural background ANC value. Some consideration of chronically acidic or highly acid sensitive surface waters that would not be recoverable at a given ANC limit should be given in the next draft of the PA.

*18. What are the views of the Panel on the use of regional air quality modeling (e.g., CMAQ) results to establish the concentration-to-deposition ratio ( $VNO_y$ ,  $VSO_x$ ) and reduced nitrogen deposition ( $NH_x$ ) in the AAPI calculation? What are the views of the Panel on the critical uncertainties associated with the use of CMAQ to generate these parameters, and the potential implications of these uncertainties for the secondary standards?*

There are a variety of problems with the use of regional air quality modeling to establish  $VNO_y$ ,  $VSO_x$  and  $NH_x$ . Although CMAQ is a regional air quality model that has been under development for many years, it has limitations in the prediction of the concentrations and deposition of different chemical species within the  $NO_y$ ,  $SO_x$ , and  $NH_x$  categories. Another issue is that there is an inherent, unvalidated assumption in establishing fixed  $VNO_y$  and  $VSO_x$ , namely that these ratios don't vary with changes in emissions.

There are many sources of uncertainties that can affect the final values derived from the model. These include, but are not limited to:

- uncertainties in emissions (particularly  $NH_x$  emissions, but also  $NO_x$  emissions);
- inability of models to correctly predict deposition from clouds and fog (both with respect to spatial distribution and liquid water content) and precipitation; and
- incomplete representation of chemistry (particularly cloud chemistry, nighttime chemistry and reactions with terrestrial components).

A comparison 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) be performed to assess the uncertainty in the model:

1. 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,

2. Model performance for wet deposition of sulfate, nitrate, and ammonium using the National Atmospheric Deposition Program (NADP) network,
3. A regional model evaluation using the continuous measurements of nitric oxide, nitrogen dioxide, nitric acid and  $\text{NO}_Y$  from the SEARCH network in the southeastern U.S.

In addition, the CMAQ model should be run with different anthropogenic emissions scenarios (keeping the other parameters and inputs constant) to quantify the sensitivity of  $\text{VNO}_Y$  and  $\text{VSO}_X$  to emissions change.

19. What are the views of the Panel regarding presentation of the standards as a set of trade-off curves for  $\text{NO}_X$  and  $\text{SO}_X$  associated with specific levels of AAPI, and specific values of the  $g$ ,  $V_{\text{NO}_X}$ ,  $V_{\text{SO}_X}$  and  $\text{NH}_X$  terms?

The Panel supports the conceptual model of the of the trade-off curves for different deposition and concentration values of  $\text{NO}_X$  and  $\text{SO}_X$  for different levels of AAPI. The Panel notes that similar curves have been developed using dynamic models. An example is shown below developed for the response of Watershed 6 at the Hubbard Brook Experimental Forest, NH to different scenarios of sulfate and nitrate deposition using the model PnET-BGC (Figure 1). The Panel urges the EPA to consider expanding this approach to also address tradeoffs associated with ammonium deposition.

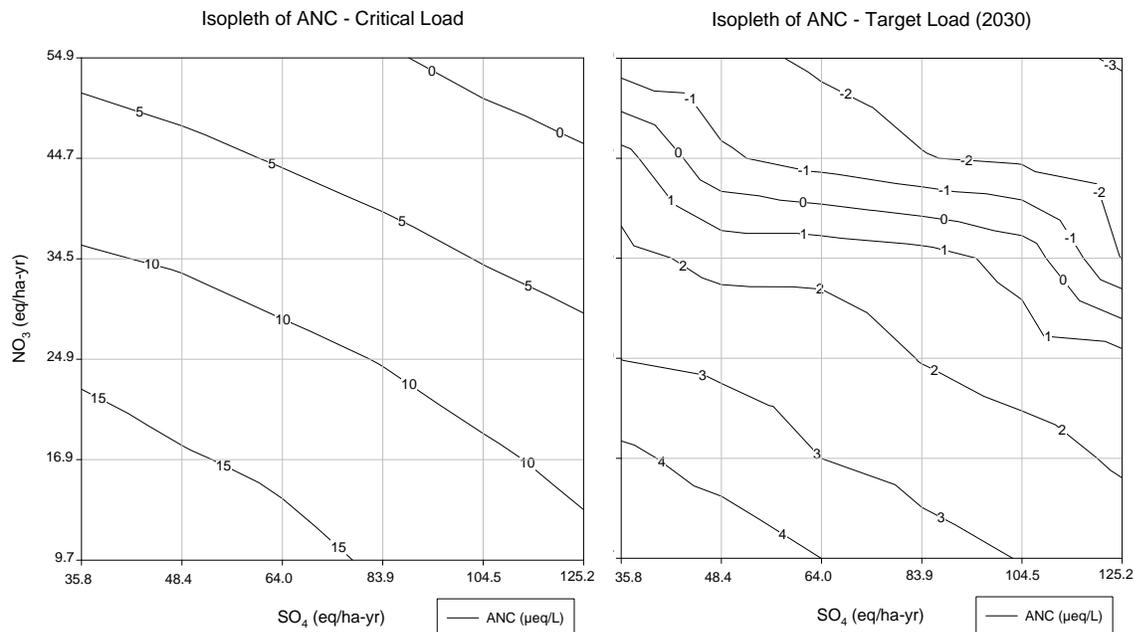


Figure 1. Response surfaces calculated for different conditions of stream ANC under different combinations of atmospheric nitrate and sulfate deposition for Watershed 6 at the Hubbard Brook Experimental Forest, NH for two periods at steady-state (~2150) (left) and at 2030 (right). The lines represent different values of predicted ANC (C.T. Driscoll unpublished).

*20. What are the views of the Panel on using a single year or a three-year average of recent year CMAQ modeling results to estimate the AAPI terms?*

The Panel supports using multi-year averages of CMAQ modeling results to estimate terms in the AAPI equation. A three-year average would help smooth out the year-to-year climatic variation in air concentration and deposition estimates. The Panel would also support four or five years.

*21. What are the views of the panel on the ambient monitoring requirements? Are the proposed three ambient air measurements – NO<sub>y</sub>, SO<sub>2</sub> and particulate sulfate – sufficient to judge compliance with and AAPI?*

The proposed measurements of ambient concentrations of SO<sub>2</sub>, particulate sulfate and NO<sub>y</sub> would constitute a minimally adequate suite of measurements for determining compliance with the AAPI. To provide continuing feedback to support refinement of, and to evaluate the efficacy of strategies to attain the AAPI, it will also be important to assure continuation of existing wet and dry deposition networks like NADP and CASTNET, to enhance those networks to add better measurements of gaseous ammonia concentration and deposition, and to assure the continuation of aquatic chemistry and effects monitoring efforts like the LTM and TIME programs. The Panel also recommends an expansion of existing programs to include some soil (especially evaluating changes in base cation status) and biological monitoring.

*22. What are the views of the Panel on using existing NO<sub>y</sub> and PM<sub>2.5</sub> sulfate measurement techniques as the basis for defining a Federal Reference Method to judge compliance with and standard?*

PM<sub>2.5</sub> sulfate measurement techniques are well established, consistently produce similar results across several different national networks, and should provide a reasonable basis for defining an FRM for compliance determination. Currently available NO<sub>y</sub> measurement techniques should also provide a sound basis for defining a FRM method. NO<sub>y</sub> measurements have been conducted successfully for many years by the research community, and are currently experiencing more widespread deployment in routine state-operated monitoring networks. The methods are robust and standardized and it's likely that early feedback from current monitoring efforts will identify any common problems with the methods and lead quickly to refined operational procedures. EPA is encouraged to consult with the CASAC AAMM Subcommittee to review options for developing standard methods for measuring SO<sub>x</sub> and NO<sub>y</sub> species for compliance determination, as well as for measuring other key species such as gaseous ammonia and organic nitrates which are currently not measured in routine monitoring networks.

## **Chapter 7: Co-protection**

*23. What are the views of the panel on the approach taken to compare the protection provided by a potential aquatic acidification standard to the protection needed for terrestrial acidification?*

The panel found it appropriate for EPA Staff to consider the potential interrelationships between aquatic acidification standards and protection of adjacent or closely related terrestrial ecosystems. Recognition that (1) various species of nitrogen (e.g., NH<sub>4</sub> or NO<sub>3</sub>) and sulfur may cycle within or move out of ecosystems differently from each other based on their chemical and physical properties; and (2) aquatic and terrestrial systems behave quite differently in the biogeochemical, temporal, and spatial processing of nitrogen and sulfur should be clearly reiterated in this discussion. Chapter 7 should be expanded to better explain the potential interrelationships between aquatic and related terrestrial responses to acidification.

*24. What are the views of the panel on a future comparison of the protection provided by a potential aquatic acidification standard to nutrient enrichment benchmarks? What are the views of the panel regarding using a nutrient enrichment benchmark to be a limiting factor on the nitrogen in the aquatic acidification standard, instead of having a separate standard?*

Dynamic models of ecological response should be considered as the primary means of addressing a continuum of responses linking terrestrial and aquatic system following nitrogen or sulfur inputs. The search for simple relationships between aquatic and terrestrial responses to regional nitrogen and sulfur atmospheric inputs is a simplification of complex processes operating on divergent spatial and temporal scales. Where sulfur is the primary acidifying agent in a given region, one should not expect to find a related response to nitrogen enrichment. It is important to recognize that the retention or lack of retention of nitrogen is controlled almost entirely by biological processes whereas the retention of sulfur is controlled by both abiotic (e.g., chemical adsorption) and biological processes (e.g., net immobilization or mineralization); thus, a combined standard in sites dominated by sulfur deposition may or may not protect from excessive nitrogen and vice-versa. The Panel is concerned that a single standard addressing both acidification and nutrient enrichment is probably not practical at this point.

## **Chapter 9: Conclusions**

*25. What are the views of the Panel on the preliminary staff conclusions regarding the adequacy of the current standards, the need for an integrated multi-pollutant structure for revised standards, and the proposed form of the joint NO<sub>x</sub> SO<sub>x</sub> standards for aquatic acidification?*

The review of current standards and the documentation of the need for an integrated multi-pollutant structure for revised standards are well done. This chapter summarizes the importance of developing new secondary joint standards for NO<sub>x</sub> and SO<sub>x</sub>. The panel supports the current focus on aquatic acidification using ANC as a key parameter. The use of the AAPI for evaluating freshwater acidification is a new and creative approach for helping the development of the joint secondary standard for NO<sub>x</sub> and SO<sub>x</sub>. The use of the BC/Al ratio in evaluating

acidification of terrestrial ecosystems appears to be an appropriate tool. Further development of a method that evaluates the impact of nitrogen as a nutrient for terrestrial and aquatic ecosystems will help expand the spatial scope of the secondary standard with respect to the effects of nitrogen deposition.

*26. What are the views of the Panel on the overall characterization of uncertainty as it relates to the determination of an ecologically-relevant multi-pollutant standard for NO<sub>x</sub> and SO<sub>x</sub>?*

As we note a number of times, the treatment of uncertainty is yet to be fully developed and the Panel recommends that this categorization be provided as a separate chapter in the next version of the report. This chapter will need to discuss the major sources of uncertainty including: 1) the estimates of the atmospheric deposition of nitrogen and sulfur; and 2) the resultant ecosystem effects caused by acidification and addition of nitrogen as a nutrient. The major sources of uncertainty will need to be highlighted in the conclusions. The listing of future research needs to be reconsidered with respect to the comments provided by the Panel so that there is a clear priority with respect to the most critical needs for further development and evaluation of secondary standards for NO<sub>x</sub> and SO<sub>x</sub>. Linking these needs to the most important aspects of uncertainty will strengthen the arguments for these research efforts.

**Enclosure C**

**Review Comments from the NO<sub>x</sub>-SO<sub>x</sub> Secondary Panel on the *Policy Assessment for the Review of the Secondary National Ambient Air Quality Standard for NO<sub>x</sub> and SO<sub>x</sub>: First Draft***

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## Dr. Praveen K. Amar

### Chapter 3: Considerations of Adversity to Public Welfare

**Charge Question 4. What are the views of the Panel regarding the characterizations of adversity to public welfare presented in the document? What are the views of the Panel regarding the use of the ecosystem services framework as an additional metric to inform questions of adversity? What are the views of the Panel regarding the usefulness of including economic valuation of some of these ecosystem services in the policy assessment document?**

Chapter 3 covers three areas of :a) adversity to public welfare, b) application of ecosystem services framework as a way/metric to address adversity to public welfare, and , c) usefulness of economic valuation approaches to “value” ecosystem services, when doable. I think that the overall effort is quite good. My main and general comment and concern is that of the three attributes of the adversity, “type, intensity, and scale,” (page 62, line 9), Chapter 3 needs to be improved with respect to explaining the *scale* of adversity to public welfare driven by “delta” or *changes* to ecosystem services as a function of *changes* in atmospheric deposition of SO<sub>x</sub>, NO<sub>y</sub>, and potentially *no changes* (potentially increases) in atmospheric deposition of reduced NH<sub>x</sub>. The Chapter presents many quantitative numbers in dollars without the proper context (at least for me) when economic valuation data are presented. I would note that the “purpose of numbers is not numbers at all but insights.” And, insights for policy makers are missing here. Also, see my response to Charge Question 5 below.

**Charge Question 5. To what extent is the presentation of the ecosystem services in the document scientifically sound and clearly communicated?**

The concept of ecosystem services and its relationship to adversity and public welfare is reasonably clear to me. However, as I note above, the communication of quantitative information can be and should be improved. One suggestion is that many “*changes*” in dollar values for ecosystem services caused by increased or decreased SO<sub>x</sub> and NO<sub>y</sub> deposition should be presented in Figures and Tables (or both) for better communication with the reader. It would also be helpful to write and comparatively explain these numbers so that the audience knows that “\$0.69 million, or only \$690,000” (value of “public welfare gains related to U.S. sugar maple and red spruce forest markets by eliminating critical load exceedances,” page 84) is quite “different” (in fact, different by four orders of magnitude or, by a ratio of 10,000 to 1) than \$11.59 billion or \$11,590,000,000 (annual benefit for California residents from trail hiking, page 88). Do these two numbers make sense in a comparative economic valuation mode? I do not know, but I am puzzled. Please present all dollar numbers with comparative insights. Otherwise, they are just numbers and not insights.

**Charge Question 6. What are the views of the Panel on the critical uncertainties associated with articulating adversity to public welfare that need to be characterized in terms of their potential implications for the secondary standards?**

It seems to me that the valuation (monetized or not) of all types of ecosystem services (provisioning, regulating, cultural, and supporting) needs to be addressed with the recognition that different levels of uncertainty are associated with valuation (monetized or not) of the four important services ecosystems provide towards public welfare.

I commend EPA for asking the question on uncertainty “correctly” in that it focuses not on the uncertainties associated with articulating adversity to public welfare themselves (“there will always be uncertainties,” what else is new ?), but on the *effect* of uncertainties on the “implications for the secondary standards.” My understanding is that the second draft of Policy Assessment will be more descriptive and more quantitative than this draft on how overall uncertainties across the whole spectrum of this approach (atmospheric and ecological models, measurements of atmospheric and ecological data, valuation of ecosystem services, etc.) will be addressed. It will be a good idea if all the uncertainties and their implications be described in one place under one chapter in some integrated and relative manner (in an Appendix?).

### **Chapter 5: Conceptual Design of an Ecologically Relevant Multi-Pollutant Standard**

**Charge Question 10. What are the views of the Panel on staff’s proposed conceptual framework for the structure of a multi pollutant, ecologically relevant standard for NO<sub>x</sub> and SO<sub>x</sub>? 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 NO<sub>x</sub> and SO<sub>x</sub>?**

The proposed 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 NO<sub>y</sub> and SO<sub>x</sub>, and surface water chemistry), with one major exception noted below.

Even though the framework and the structure “takes into account” the reduced ambient NH<sub>x</sub> 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 national ammonia emissions under proposed standard setting structure. Ammonia emissions are currently at about 4 to 5 million tons per year, and are rising. As the Executive Summary notes on Page ES-7, this approach assumes that “the reduced forms of nitrogen deposition are relatively constant over time. This assumption could lead to over or under protection for an area depending on whether the actual concentrations of reduced forms of nitrogen increase or decrease over time.” For the case of unregulated ammonia as a criteria pollutant at the present time, emissions (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 CAFO sources (confined animal feeding operations) in the U.S.

Notwithstanding my concern about not addressing reduced nitrogen directly, the proposed structure more than adequately represents the scientific linkages between ecological effects, surface water chemistry, atmospheric deposition, and ambient levels of NO<sub>y</sub> and SO<sub>x</sub>.

**Charge Question 11. What are the views of the Panel on the *relevance* of the conceptual design for developing standards to protect against aquatic acidification effects?**

**Charge Question 11. a. What are the views of the Panel on staff's proposed options for the ecosystem acidification model (s) to represent the ecological response function?**

I do not have background or expertise in the design and application of ecosystem acidification models (MAGIC, FAB, SSWC, etc.) used in this Policy Assessment and I found it difficult to follow the general flow of discussion. A general comment that can be made about these models is that they need to be "validated/verified/calibrated" to increase confidence in their results. This Chapter notes that model such as MAGIC, "because of their aggregated nature, need to be calibrated to observational data from a system before it can be used to examine potential system response." A large set of observational data on stream water quality, soil chemical and physical characteristics, etc., are needed. My general question is on the degree and extent of evaluation of ecological acidification models compared to, say, the evaluation/calibration of atmospheric models such as CMAQ. For example, recognizing that these models do involve different physical, chemical, and biological systems, is the level of confidence in these models at a "comparable" level?

**Charge Question 11. b. What are the views of the Panel on the relative merits of the two techniques for calculating Neco, the parameter representing the amount of deposited nitrogen that is available for acidification due to uptake, denitrification and immobilization?**

As in above question, this is not my area of expertise and I am not sure which approach is more reliable. It appears to me, however, that the second approach (equation (4)), based on measured N leaching in a catchment, and may be more accurate. Is it? Is it also more protective? The text (page 160) briefly (but, not clearly) describes a *third* approach where SSWC results on critical load are constrained by Neco (how and by how much?). Which of the three approaches provide the optimum balance between accuracy/reliability and ease of use? I assume our ecology colleagues will shed more light on this question.

**Charge Question 11. c. What are the views of the Panel on staff's proposed options for developing acid sensitivity classes to categorize the national landscape? Is it appropriate to base this classification on bedrock geology? Should multiple criteria be used to inform the sensitivity categories?**

The literature and published research in the field indicates that bedrock geology is an acceptable basis for developing acid-sensitivity classes on the larger scale of the U.S. One way to answer the question whether other multiple features/multiple criteria, in addition to bedrock geology, may/should be used as the basis for national categories of acid-sensitivity would be to do a "sensitivity analysis." This could involve other variables to which acid-sensitivity has been

shown to be correlated with (for example, elevation, watershed area, percent forested watershed, etc.). However, this type of investigation needs to be well-designed and focused, taking into account co-variables and would need to be validated with actual field data (for example, with surface water chemistry data) before multiple criteria are selected to redefine acid-sensitivity categories on the national scale,

**Charge Question 11. d. What are the views of the Panel on staff's proposed options for how to aggregate acidification modeling from the catchment-scale to represent acid-sensitivity categories at the national-scale?**

Of course, this question is on how to “add up” results of acidification modeling at catchment-scale to represent acid-sensitivity categories at the national scale. As the Policy Assessment notes, the proposed standard would be designed such that “a specified percentage of a population of water bodies does not exceed a critical load for the specified value of ANC.” What is not clear is how this percent of lakes/streams would be chosen. For example, would one choose, say, 50% of lakes or 95% of all lakes to be protected? I assume it is a policy call of EPA. This also implies that this percent number will be chosen for a certain area of certain spatial extent/size (all of Adirondack? Adirondack subdivided by geologic characteristics? by elevation?). The issue here is the difficulty of deciding an acceptable degree of representativeness of a small sample of measured lakes to represent the whole population of lakes/streams with a large acid-sensitivity distribution. One potential and, may be better, approach would be to only include those lakes in the sample and in the total population that, based on current and/or historic conditions, are considered to have high level of sensitivity to acidification in the first place. This is analogous to establishing primary (public health) NAQQS, based on protecting the most sensitive fraction of human population.

**Charge Question 11e. What are the views of the Panel on staff's suggested method to account for reduced nitrogen in the deposition metric?**

As I note in response to Charge Question 10, even though the framework and the structure of proposed approach “takes into account” the reduced ambient NH<sub>x</sub> and its deposition in designing AAPI (atmospheric acidification potential index), it does so in a manner such that it most probably will not allow EPA to address and require reductions in national ammonia emissions under the proposed standard structure. Ammonia emissions are currently at about 4 to 5 million tons per year and, unlike SO<sub>x</sub> and NO<sub>x</sub> emissions, are rising. As the Executive Summary notes on Page ES-7, this approach assumes that “the reduced forms of nitrogen deposition are relatively constant over time. This assumption could lead to over or under protection for an area depending on whether the actual concentrations of reduced forms of nitrogen increase or decrease over time.” For the case of unregulated ammonia, emissions (and resulting ammonia and ammonium concentrations and deposition of reduced nitrogen) are only expected to increase by as much as ten percent over the next few decades because of higher human population, increased food production and increased CAFO activity (confined animal feeding operations) in the U.S.

**Charge Question 12. What are the views of the Panel on the relevance of the conceptual design for developing standards to protect against terrestrial acidification effects? terrestrial nutrient enrichment effects? aquatic nutrient enrichment effects? Does the Panel**

**have suggestions on additional data or methods that might enable EPA to expand the current aquatic acidification approach to cover additional effects?**

The design approach developed here for aquatic acidification should be relevant to developing future standards for terrestrial acidification effects. The “BC/Al” ratio appears to have the same level of robustness as an ecological indicator as ANC is for lakes and streams. It meets the important criterion that it can be correlated with ecological effects on forests and soils, and also through models, to acid deposition levels. As the Assessment notes, in general, the aquatic critical loads offer greater protection to the watersheds than do the terrestrial critical loads. However, this relationship changes depending upon whether lakes are of “low sensitivity”, “not sensitive”, “highly sensitive” or “moderately sensitive” etc. Therefore, it would be useful for EPA to allow sufficient flexibility into the process of expanding the approach outlined here to terrestrial acidification, as well as to other ecological effects. For the case of nutrient enrichment of coastal estuaries, it would be extremely useful to increase the level of confidence (through additional modeling and measurements) in determining the fraction of reactive N (including reduced nitrogen, NH<sub>x</sub>) loading that is attributable to atmospheric deposition.

**Charge Question 13. What are the views of the Panel on the critical uncertainties associated with the conceptual design of an ecologically relevant multi-pollutant standard that need to be characterized in terms of their potential implications for the secondary standards?**

This is important question and, equally important, is asked “correctly” in that it focuses not on the uncertainties themselves (“there will always be uncertainties”) but on the *effect* of uncertainties on the implications for setting the secondary standards. My understanding is that the second draft of Policy Assessment will be more descriptive and more quantitative than this draft on how uncertainties across the whole spectrum of this approach (atmospheric and ecological models, measurements of atmospheric and ecological data, ecosystem services) will be addressed. It will be a good idea if all the uncertainties and their implications be described in one place under one chapter in some integrated and relative manner (in an Appendix?). The discussion of “uncertainties” in this draft (in this Chapter and throughout the report) is generally descriptive in nature. It is, however, not helpful in providing a quantitative and relative sense of uncertainties in various components (emissions, wet and dry deposition predictions and measurements, ecological models versus CMAQ, etc.). For example, it would be useful to state that NH<sub>3</sub> emissions are much more uncertain than NO<sub>x</sub> emissions that in turn are less certain than SO<sub>2</sub> emissions. It may also be useful to state that dry deposition is simply not measured but inferred from model calculations. For variables that have uncertainties of unknown magnitude, sensitivity studies could be useful to put bounds on the results.

**14. To what extent do the figures and examples aid in clarifying the text? Should more or less information of this type be included in the second draft?**

The Figures, Tables, and examples were very useful in understanding the text material, especially when subject discussed was not in my area of expertise (ecological models). If anything, I would have preferred more Tables, Figures, and examples to explain abstract ideas in the text.

### **General Editorial Comments:**

Page 147, Line 10: I think you need to say not a DESIRED level but a “SPECIFIC or GIVEN” level.

Page 147, Lines 21, 23, please avoid “Let us now.”” Or “ you could calculate..”

Page 148, Figure 5-2 Title: It is too long, and needs to be shortened and clarified.

Page 148, Line 7-8: Please restate, “it is not practical to evaluate each catchment individually since it is not the appropriate approach for a national standard.”

Page 148, Line 11; “.. as far as the relationship of total deposited N and S to ANC IS CONCERNED.”

Page 148, Line 18: Please explain here what is meant by  $DL_{\%ECO}$ ?

Page 149, Line 28: please explain here what is meant by “recently developed base cation surplus.....”

Page 151, Line 2-4: Please write clearly to say “.. show long-term trends in RELATIONSHIP between anthro N and S dep and ANC .....”

Page 151: Table should be labeled Table 5.2-1. Also, the accompanying text needs to explain how “single value” is different from critical load function.

Page 151, Line 16: Please describe and compare and contrast the two approaches mentioned here: time series analysis and critical load approach

Page 151: Line 12-13 The Title 5.2.3 needs to say “... indicator AND amount of N and S deposition.”

Page 152, Line 30: “.. the relationship BETWEEN ANC and deposition.”

P 154, Line 10: “Consideration of the effects.... is REQUIRED extensively.....”

P 154: Lines 20 and 29; please be parallel and use the term “surface water quality data”

P 155, Line 28: The technique does not DETERMINE the number of streams, it simply estimates the number of streams.

P 156, Line 15: ‘the relationship BETWEEN ANC and deposition

Page 161, Line 17: “...this technique is SHOWN...”

## Dr. Andrzej Bytnerowicz

### Chapter 6: Options for Elements of a Standard to Protect Against Effects from Aquatic Acidification

**15. To what extent does the Panel agree that the proposal to develop an Atmospheric Acidification Potential Index (AAPI) linked to key determinants of aquatic ANC is a reasonable approach to developing an ecologically relevant standard? Does the Panel generally agree that the secondary standard options identified by staff (including indicator, averaging time, form, and level) are generally consistent with the available scientific and technical information and are appropriate for consideration by the Administrator?**

In a general sense this is a reasonable approach that links ecological responses (ANC as an ecological indicator), acidifying deposition of N and S (resulting from most important forms of atmospheric sulfur and reactive nitrogen), and ambient NO<sub>x</sub> and SO<sub>x</sub> concentrations. Science and logic behind this concept are good and based on the current understanding of the effects of atmospheric deposition on aquatic ecosystems. I would like to complement the EPA authors for developing a concept that logically links the concentration-based standards for NO<sub>x</sub> and SO<sub>x</sub> with ecological effects of total N and S deposition that at the same time includes reduced form of N as part of the background chemical environment of the evaluated watersheds. Potential problems related to the proposed approach are mainly related to conversion of deposition to concentrations and to the unknown levels of uncertainty of various stages of the proposed methodology. While an averaging time of a single year for acidification of aquatic ecosystems could be probably justified, a similar approach for the terrestrial ecosystems is too coarse and therefore is not recommended. For terrestrial ecosystems the annual averages should be based on aggregated values of the individual phases of physiological activity of vegetation specific for various ecological zones.

**16. What are the views of the Panel on the degree of uncertainty associated with each element of the suggested standard, e.g. the ecological indicator; the concentration to deposition ratios, reduced nitrogen, the natural background ANC, and the ambient indicator and averaging time for NO<sub>x</sub> and SO<sub>x</sub>, and its relationship to the degree of protection that could be expected from the standard? What are the views of the Panel on how to fairly characterize the uncertainty associated with the degree of protection that such a standard would provide for aquatic acidification?**

Scientific information on levels of acid neutralizing capacity (ANC) protective to fish and aquatic biota has been well developed and therefore present the lowest level of uncertainty. On the other hand, the concentrations to deposition ratios present a very high level of uncertainty. Various individual components behind the total V<sub>N</sub> or V<sub>S</sub> are not very well defined and could be quite variable. According to my understanding of the problem, a level of uncertainty for water surfaces is lower than for the terrestrial ecosystems. Deposition velocities of reactive N species, such as NH<sub>3</sub> and organic N, both for the aquatic and terrestrial ecosystems have high level of uncertainty. Information on ambient levels and distribution of the reduced N forms, NH<sub>3</sub> and

$\text{NH}_4^+$ , are scarce and therefore there is very high level of uncertainty on distribution of these pollutants, especially in remote areas, and specifically in the mountainous complex terrain. Ammonia concentration values from the CMAQ model are not reliable and have not matched the ground-level measurements (Fenn et al., 2009). In summary, the expected level of uncertainty of the proposed methodology for protection against acidification of the aquatic ecosystems is high.

**17. What are the views of the Panel on aggregating the terms ( $Q$ ,  $N_{\text{eco}}$  and  $\text{BC}_0^*$ ) used in estimating natural background ANC, denoted by the function  $g(\cdot)$ , into 5 bins based on the geologic classification scheme?**

Introduction of function  $g(\cdot)$  aggregating  $Q$ ,  $N_{\text{eco}}$  and  $\text{BC}_0$  seems to be appropriate. Using the geologic classification scheme is a good idea since chemistry and weathering of geologic material are the main factors affecting abilities of the aquatic ecosystems to cope with acidification caused by the atmospherically deposited S and N compounds.

**18. What are the views of the Panel on the use of regional air quality modeling (e.g., CMAQ) results to establish the concentration-to-deposition ratio ( $V_{\text{NO}_y}$ ,  $V_{\text{SO}_x}$ ) and reduced nitrogen deposition ( $\text{NH}_x$ ) in the AAPI calculation? What are the views of the Panel on the critical uncertainties associated with the use of CMAQ to generate these parameters, and the potential implications of these uncertainties for the secondary standards?**

First, I would like to suggest changing symbol “V” describing “concentration-to-deposition ratios” to a different one. Symbol “V”, at least in my understanding, has been used for description of deposition velocity (calculated by dividing deposition, or flux, of a given pollutant by its ambient concentration). As I stated above (point No. 16), the CMAQ model outputs often do not agree with the ground-level measurements, especially for the reduced N compounds (Fenn et al., 2009). Therefore I strongly support the recent EPA’s efforts to enhance a national network of  $\text{NH}_3$  monitoring using passive samplers. This effort should continue especially in remote areas. Implementation of passive sampling methodology is needed and various passive samplers (such as Alpha, Ogawa or Radiello) could be used. It would make sense to include such measurements as a permanent feature of the national monitoring networks such as CASTNET or NADP.

**19. What are the views of the Panel regarding presentation of the standards as a set of tradeoff curves for  $\text{NO}_x$  and  $\text{SO}_x$  associated with specific levels of AAPI, and specific values of the  $g$ ,  $V_{\text{NO}_x}$ ,  $V_{\text{SO}_x}$ , and  $\text{NH}_x$  terms?**

It is a reasonable way of dealing with two elements which simultaneously are responsible for acidification of the aquatic and terrestrial ecosystems. As I have stated above, the V values for specific components of deposition generally will have a high level of uncertainty. Therefore final calculations of the  $\text{NO}_x$  or  $\text{SO}_x$  concentrations might be problematic.

**20. What are the views of the Panel on using a single year or a three year average of recent year CMAQ modeling results to estimate the AAPI terms?**

A single year or an average of three years of measurements based on the CMAQ model is not adequate. There will be large differences in concentrations of the pollutants responsible for acidification as well as significant spatial and temporal differences in N and S fluxes due to changing weather, distribution of emission sources, and levels of emissions. With an increasing use of green energy and unpredictable climate changes, year-to-year information on annual values (possibly also presented as a running mean of three years) would greatly help in reliable estimates of the AAPI values and reduction of their uncertainties.

**21. What are the views of the panel on the ambient monitoring requirements? Is for the proposed three ambient air measurements – NO<sub>y</sub>, SO<sub>2</sub> and particulate sulfate – sufficient to judge compliance with and AAPI?**

Some of the major components of NO<sub>y</sub> are measured on national air quality monitoring networks (NO, NO<sub>2</sub> and SO<sub>2</sub>). Fine particulate sulfate has been extensively measured within the IMPROVE and CASTNET networks in remote areas of the US. Among the NO<sub>y</sub> species, HNO<sub>3</sub> and particulate NO<sub>3</sub> are measured in the CASTNET network. However, organic N is not routinely measured although it may provide substantial levels of reactive N (Zhang et al, 2008). In summary – for a successful national program designed for a long-term understanding of acidifying effects of atmospheric deposition, and information on relevant N and S species, a thorough evaluation of the national air quality monitoring efforts in remote areas is needed. Some techniques, such as the already mentioned passive sampling, should be considered. They can be successfully used for monitoring concentrations of NO, NO<sub>2</sub>, NH<sub>3</sub>, HONO and HNO<sub>3</sub> and SO<sub>2</sub>. Such efforts are already underway in Europe on the ICP Forests Level II plots and have also been conducted in selected areas of the United States and Canada.

**22. What are the views of the Panel on using existing NO<sub>y</sub> and PM<sub>2.5</sub> sulfate measurement techniques as the basis for defining a Federal Reference Method to judge compliance with and standard?**

Use of chemiluminescence for total NO<sub>y</sub> measurements seems to be an accepted methodology although uncertainties exist related to reduction of the organically bound and mineralized N (page 208 of the reviewed document and Navas et al., 1997). Extensive sulfate measurements have been conducted nationwide within the CASTNET, IMPROVE and EPA CNS networks. It seems that these methodologies could be used for defining the Federal Reference Method, although an opinion of experienced atmospheric chemists is recommended.

References:

Fenn, M.E., Sickman, J. O., Bytnerowicz, A., Clow, D. W., Molotch, N. P., Pleim, J. E., Tonnesen, G. S., Weathers, K. C., Padgett, P. E., Campbell, D. H. 2009. Methods for Measuring Atmospheric Nitrogen Deposition Inputs in Arid and Montane Ecosystems of Western North America. *Developments in Environmental Science*, Vol. 9, Allan H. Legge (Ed.), 177-225.

Navas, M. J., Jimenez, A. M., Galan, G. 1997. Air analysis: determinations of nitrogen compounds by chemiluminescence. *Atmospheric Environment*, 31, 3603-3608.

Zhang, Y., Zheng, L., Liu, X., Jickells, T., Cape, J. N., Goulding, K., Fangmeier, A., Zhang, F. 2008. Evidence for organic N deposition and its anthropogenic sources in China. *Atmospheric Environment*, 42, 1035–1041.

## Ms. Lauraine Chestnut

### Chapter 3: Considerations of Adversity to Public Welfare

**Charge Question 4: What are the views of the panel regarding the characterization of adversity to public welfare presented in this document? What are the views of the panel regarding the use of the ecosystem service framework as an additional metric to inform questions of adversity? What are the views of the panel regarding the usefulness of including economic valuation of some of these ecosystem services in the policy assessment document?**

Chapter 3 does a nice job at the beginning in bringing in language from other agencies and other regulatory assessments to articulate why protection and preservation of natural environmental resources are important and motivate legislation and regulation in many areas. I especially like the words from the FWS about wildlife refuge systems being managed to “ensure that the biological integrity, diversity, and environmental health of the Systems are maintained for the benefit of present and future generations of Americans.” (underline added)

The focus in the text is on federal protections, but it is important to recognize that the public cares about all kinds of natural areas including state and local parks as well as the federal Class I areas. This distinction is important for the management authorities, but not so much for the public. While Figures 3-4 and 3-5 have the right idea in terms of concept, the focus on NPS Class I areas is too limited. There are many areas throughout the country that are largely natural where people drive, fish, hike, sight see and enjoy the natural flora and fauna, including state and local parks, national and state forests, and all the lakes and streams therein. Even though these do not have the protection levels of the national parks and wilderness areas, and there may be multiple uses such as timber harvesting and grazing, the value of these areas stems largely from their natural characteristics and the public cares about the “integrity, diversity and environmental health” of these areas.

The basic concepts of ecosystems services are useful to explain why the health of these systems matters. To aid this process of providing information to help the Administrator assess adversity with regard to determining an appropriate level for the secondary standard, more is needed than just a conceptual framework. The framework needs to be used to characterize the change or loss in services that currently exists as a result SO<sub>x</sub>/NO<sub>x</sub> deposition. Chapter 3 does a reasonable job with this for the aquatic acidification case, but not as well for the others. It is ideal if this change or loss can be defined quantitatively, but this is not necessary. The change or loss can be described based on causal linkages that are well documented even if they cannot be quantified with specific dose-response relationships.

The economic valuation estimates included in Chapter 3 are useful when they are tied to the change or loss in value of ecosystem services that can be reasonably attributed to SO<sub>x</sub>/NO<sub>x</sub> deposition. Total values of services that flow from the effected ecosystems are less useful because they convey little about what share of this value is being lost. In fact, if they are based

on current service levels then they are already missing the value of what has been lost due to harmful effects of SO<sub>x</sub>/NO<sub>x</sub> deposition.

Page 76: The discussion here unnecessarily undercuts the usefulness of economic valuation estimates. It is true that just because there is some quantifiable economic welfare loss does not mean that the loss is necessarily significant enough to be adverse. However, demonstrating a substantial loss in economic valuation terms would provide strong support for a determination that there is an adverse effect, even though the numbers alone are not enough to specify the specific threshold at which a loss become substantial rather than trivial.

Pages 77-78: The comments here about the assumptions in economics that “public preference are paramount” are misleading. Economic valuation takes expressed preferences to be a valid measure of welfare when they are based on accurate information. If people do not express a value to protect ecosystems from the effects of pollution because they do not understand the implications of these effects, then such lack of value should not guide policy decisions.

Page 80: I think the Banzhaf et al. (2006) study should be given more mention than only in footnote 11. It provides strong evidence that the public cares about water quality in Adirondack lakes being sufficient to support native species, even if they do not participate in fishing. It is important to emphasize that these results are only for residents of NY state. Residents in other states could also be expected to value water quality in the Adirondack area. EPA may not have a great deal of confidence in the specific quantitative estimates based on extrapolations from the Banzhaf et al. study, but the fact that the results exceed by a large amount the estimates of the value of lost recreational fishing services is a very important finding that should be emphasized.

Page 82: The estimates of economic value of lost recreational fishing services in the Adirondack region are not very well explained. The \$4 million is the annualized value of reaching an ANC level of only 20. At ANC of 50 or 100, the annualized value is \$8 to \$9 million. This seems like the most relevant range. The \$300 million number is a present value through 2100. (REA, Appendix 8, Table 2.2-6) When the fishing losses are extrapolated to all the comparable lakes in NY state, the estimate of annual value of gaining ANC of 50 is \$25 to \$28 million, and of gaining ANC of 100 is \$110 to \$130 million, and present values are in the billions (REA, Appendix 8, Table 2.2-7). It is not enough to just say that the range is \$4 million to \$300 million. The numbers need a bit more explanation. It is also not clear what point is being made in lines 11-15. In contrast to these numbers, the estimates of annual value to all NY state households of reducing acidification in the Adirondacks region to an ANC of 50 is \$300 million to \$800 million based on extrapolations from the Banzhaf et al. study (REA, Appendix 8, Table 2.2-9). These estimates are worth mentioning in Chapter 3. They reflect use and nonuse values held by the public for protecting the area and the underlying study specifically described the effects of acidification in the region.

The discussion of the terrestrial effects of acidification would benefit from a descriptive statement about how the terrestrial ecosystem services are affected by SO<sub>x</sub>/NO<sub>x</sub> deposition related acidification at current levels. Can something be said about forest decline? Mortality rates of trees in sensitive species in affected areas? Geographic areas affected? Visual aesthetic effects for visitors? Habitat losses or quality degradation? The statements should not be that

acidification could cause these kinds of effects, but rather, that such and such effects are documented in specific locations at current conditions.

Page 83, lines 20-28: These seem like important studies. How do the descriptions of the effects on the forests that are being valued in these studies compare to current documented effects of acidification?

Page 84: I think these are estimates of loss in commercial value of timber harvested for red spruce and sugar maple trees. It appears to be relatively small compared to the total market (of these species alone or of all timber in the region?), but this is not entirely clear. These numbers need a bit more explanation and context. They appear to show that the commercial value of timber losses for these species is relatively inconsequential.

This section is challenging because the economic valuation estimates are limited and cover only a portion of the ecosystem services affected by deposition. The bottom line is therefore irrelevant, so it is insufficient to just list various dollar amounts. It is important to include discussion of what the available estimates cover and why they are relevant—what do they say about the significance of the welfare effect? It is also important to remind the reader of the lost or impaired services that are not included in these estimates.

**Charge Question 5: To what extent is the presentation of ecosystem services in this document scientifically sound and clearly communicated?**

The basic concepts of ecosystem services and how they relate to ecosystem function and to the idea of welfare effects as defined in the CAA are clearly communicated and well presented. Description and summarization of how these services are being affected by the effects of SO<sub>x</sub>/NO<sub>x</sub> deposition need to be improved. The REA and ISA provide support for descriptions of these effects on services even though specific quantification is not feasible.

**Charge Question 6: What are the views of the panel on the critical uncertainties associated with articulating adversity to public welfare that need to be characterized in terms of their potential implications for the secondary standards?**

It is pretty clear that current levels of acidification are causing adverse effects on aquatic resources in some locations. It seems there is plenty of support to argue that it is adverse to public welfare to have a significant share of lakes and streams that cannot support several species of fish that would have lived there but for the effects of manmade pollution. Important uncertainties include how much of the adversity would be reduced with a standard based on ANC 50 versus ANC 100. The economic valuation estimates show additional value in going from 50 to 100, but they cannot give a specific estimate of how much is enough to protect public welfare. Related to this is the question of what percentage of lakes and streams need to achieve these levels to eliminate an adverse effect. The evidence may not give a definitive answer to this question, but it needs to be addressed.

**Chapter 9: Conclusions**

**Charge Question 25: What are the views of the panel on the preliminary staff conclusions regarding the adequacy of the current standards, the need for an integrated multi-pollutant structure for the revised standards, and the proposed form of the joint NO<sub>x</sub> SO<sub>x</sub> standards for aquatic acidification?**

The case has been well established that the current standards are not adequate to protect ecosystems from harmful effects of NO<sub>x</sub> and SO<sub>x</sub> deposition. The only aspect not addressed is that current exceedences of PM and ozone standards will require further reductions in NO<sub>x</sub> and/or SO<sub>x</sub> precursors as these also contribute to formation of PM and ozone. How far meeting these standards will go toward protecting ecosystem resources from effects of NO<sub>x</sub> and SO<sub>x</sub> deposition has not been addressed, and it may be quite difficult to do so.

The proposed form of the joint NO<sub>x</sub> and SO<sub>x</sub> standard makes sense conceptually. The execution of the specifics still seems problematic in several regards.

**Charge Question 26: What are the views of the panel on the overall characterization of uncertainty as it relates to the determination of an ecologically-relevant multi-pollutant standard for NO<sub>x</sub> and SO<sub>x</sub>?**

Page 220, bullet (3): The conclusions stated about limited confidence in relating natural habitat provision and biological control services to ANC levels are too cautious. A clear quantitative relationship between ANC concentrations and loss in fish species has been established. This means there is a loss in these services even if there is not a specific metric to quantify the total welfare value of this loss.

Page 220, bullet (4): Please note that the dollar values for recreational fishing losses are in the Adirondack region (or for all of NY state, depending on the numbers presented) and are for NY residents only. Lines 24-25 seem to imply they are for the whole Northeast. Note the comment on Chapter 3 about the problem with lumping annualized and present value numbers into the same “range.”

Page 221, lines 12-14. It seems appropriate to also list the recreation and aesthetic value of forest health as a type of cultural service that is harmed by the effects of acidification.

**Other important uncertainties**

Extrapolations from catchment or watershed specific analyses to regional areas are necessary to implement the proposed standards. Are these extrapolations sufficiently accurate or reliable for use in a standard setting and enforcement process? (Chapter 5, section 5.2.1.7)

Selecting a method to estimate the amount of N that will be taken up by the ecosystem seems like an important unresolved issue. (Chapter 5, page 160).

Is there any information to help guide the Administrator about what percent of an ecosystem would be appropriate for achieving a selected ANC? Are there practical constraints on what can be reasonably achieved? Is there a tradeoff, in practice, between the selected ANC target and the percentage of the area expected to meet that target?

The exclusion of reduced forms of N from the standard still seems problematic. It assumes that the contribution of these to the ecosystem effects remains the same over time. (Chapter 5, page 165).

The benchmarks for terrestrial acidification effects of 0.6, 1.2, and 10 are mentioned several times. Do these relate to lower and higher levels of effect or are these a range of estimates for a “no effects” or safe level? How far will protection of aquatic resources go toward protecting terrestrial resources and how does uncertainty about the benchmark make it difficult to answer this question?

The analysis for the acidification effects reveals a tradeoff between SO<sub>x</sub> and NO<sub>x</sub> to achieve the same target (ANC level), but this does not account for the fact that NO<sub>x</sub> contributes more to nutrient enrichment than does SO<sub>x</sub>. Thus, a comprehensive standard would take account of both types of effects. This may not be feasible at this time, but it is a drawback of focusing only on acidification in setting a standard that should be acknowledged.

## **Executive Summary**

The executive summary does a good job of summarizing the main document, but suffers from the same limitations as are in the main document. It is generally well written and easy to understand for a non-technical audience. In reviewing the ES I was looking for succinct descriptions of what the harmful effects of deposition are and to what extent are these occurring now in the US. Pages ES-3 to ES-5 are close to accomplishing this, especially for acidification effects on aquatic ecosystems. Things that still need to be explained are: (1) What does it mean in terms of the health of the trees, forest, habitat, etc. if sugar maple or red spruce growth is reduced by 20%? (2) What is the evidence and extent of observed harm to forests (or specific species) at current deposition levels? Making the link between the Bc/Al ratio and the percentage reduction in growth is very helpful for the policy analyst, but it doesn't go far enough in explaining why this matters.

At the top of page ES-5: Is there a better word than “complicated”? The point is that there are multiple sources so there is difficulty in specifying or quantifying the amount of harm done by the one source of interest here, which is atmospheric deposition.

Page ES-6: Be careful to use the term “critical load” in a consistent manner. The PA document has defined critical load as the amount of deposition that the system can tolerate and still maintain a given level of functioning, as indicated by a measure such as ANC. Thus, there is a critical load for ANC = 50 and a different critical load for ANC = 100. This means there is not one single “critical load” that determines what the policy objectives are.

Pages ES-8 and ES-9: The AAPI equation needs more explanation, term by term, for a nontechnical audience to comprehend. What is here is a good start.

Page ES-12, paragraph 4 that starts “ANC levels in the range...” This is an excellent summary of the effects on aquatic ecosystems at various ANC levels. This discussion needs to take the next step to describe what the welfare implications of these effects are.

Page ES-14: Bullets 3 and 4 are too limited on use type welfare effects. Recreational fishing and timber production are important services, but they are not the main reasons why people care about protecting these ecosystems.

## **Dr. Ellis Cowling**

My individual comments on the March 2010 First External Review Draft of the Policy Assessment for the Secondary National Ambient Air Quality Standards for Oxides of Nitrogen and Oxides of Sulfur are organized below in response to each of the several Charge Questions posed in Lydia Wegman's memoranda to Kyndall Barry dated March 4, 2010 in preparation for the April 1-2, 2010 CASAC meeting. As requested by Chairman Ted Russell, my attention has been given primarily to the three Charge Questions on Chapter 2 and the several Charge Questions on Chapter 6.

### **Overview Comments on the Whole of this First Draft Policy Assessment Document**

Before plunging directly into my assigned chapters (Chapters 2 and 6), permit me to suggest that a very constructive and useful addition to the Second Draft Policy Assessment document would be a short but very frank description of the reasons why identifying ammonia and ammonium ion (NH<sub>x</sub>) as a seventh Criteria Pollutant would not be a desirable alternative to the clever way that chemically reduced forms of nitrogen have been built into the "as given" characteristics of the various regions of our country where NH<sub>x</sub> deposition is a very significant contributing cause of both acidification and nitrogen enrichment of terrestrial and aquatic ecosystems.

This suggestion is especially relevant in view of the very thorough attention that was given to chemically reduced forms (NH<sub>x</sub>) of acidifying deposition in both:

- The ISA and the REA for the present Integrated Review of the NO<sub>x</sub>/SO<sub>x</sub> NAAQS Secondary Standards, and
- The thorough exploration of the importance of NH<sub>x</sub> as well as NO<sub>x</sub> and NO<sub>y</sub> in the soon to be completed Final Report of the Integrated Nitrogen Committee of EPA's Science Advisory Board.

### **Chapter 2: Known or Anticipated Ecological Effects**

**1. What are the views of the Panel on the appropriateness of staff's weight-of-evidence approach which assesses information from across the various ecological research areas described in the NO<sub>x</sub> SO<sub>x</sub> Secondary Standards Integrated Science Assessment (ISA), including studies of acidification and nutrient-enrichment in aquatic and terrestrial ecosystems, and laboratory research on responses of plant and animal species to acidification and nutrient enrichment? To what extent is the presentation of evidence drawn from the ecological effects studies assessed in the ISA technically sound, appropriately focused and balanced, and clearly communicated?**

The summary of "known" effects of atmospheric-deposition induced acidification and nitrogen enrichments effects presented in Chapter 2 is outstanding well done and faithful to the findings of causality and other atmospheric-ecosystem linkage relationships that are presented in the ISA and the REA reviewed earlier by the CASAC Integrated NO<sub>x</sub>/SO<sub>x</sub> Secondary NAAQS Review Panel. By contrast, "anticipated" effects are given very short shrift in Chapter 2.

I was especially delighted to see for the first time in my career -- the extraordinarily useful overview figures shown in Figures 2.8 and 2.9 on pages 42 and 43. There is no general reference as to the source of either of these two figures. Thus, I presume that they were assembled by one or more EPA staff or consultants whom I recommend develop and publish these two figures in one or more of the peer-reviewed journals of both ecology and atmospheric science. In doing so, however, it would be essential to include each of the original articles that are cited in Figure 2.8 but 17 of which are not included in the reference list on pages 52-61 at the end of Chapter 2.

My principal suggestions for improvement of this otherwise excellently well written Chapter 2 are the following:

- a) Add to the list of references on pages 51-61 all those references that are included in the body of Figure 2.8 on page 42 but are not included in the references listed at the end of the chapter.
- b) Add literature citations to the various (1-20) geographical areas where effects of acidification have been observed in Figure 2-9.
- c) Revise the caption for Figure 2.8 so it is more fully descriptive of the information contained in this figure.
- d) Revise the wording of various paragraphs within Chapter 2 to include the phrase “total reactive nitrogen,” “total reactive N,” or at least “total N” in place of just “N” as used in many places in Chapter 2 so there is no question about whether the term “N” in this chapter includes both chemically reduced as well as chemically oxidized forms of total reactive N.

This very same point is explained very thoroughly in Chapter 8 and also very well on lines 21 through 25 on page 37. But the just “N” terminology is adopted in most parts of Chapter 2. Examples of places where this suggested revision would increase the clarity of communication throughout Chapter 2 include: lines 4 and 19 on page 35, line 17 on page 37, lines 8 and 23 on page 38, lines 13 and 20 on page 39, among many others.

I also recommend that this recommended revision of wording -- including the phrase “total reactive nitrogen,” “total reactive N,” or at least “total N” in place of just “N” -- be examined not only in my two assigned chapters (2 and 6) but also in all the other chapters of this First Draft Policy Assessment document -- Chapters 3, 4, 5, 7, 8, and 9.

## **2. To what extent are the interpretation and presentation of the results of the exposure and risk assessment technically sound, appropriately balanced, and clearly communicated?**

As indicated in the first paragraph of my comments to Charge Question 1, above: “The summary of “known” effects of atmospheric-deposition induced acidification and nitrogen enrichments effects presented in Chapter 2 is outstanding well done and faithful to the findings of causality and other atmospheric-ecosystem linkage relationships that are presented in the ISA and the REA.

**What are the views of the Panel on the critical uncertainties associated with the risk and exposure analysis and the evidence from ecological effects studies that need to be characterized in terms of their potential implications for the secondary standards?**

Critical uncertainties are hardly touched on at all in Chapter 2 but seem to have been dealt with more thoroughly (and I think appropriately) in Chapters 4 and 6. Thus I refer this question also to the attention of the other CASAC Review Panel members Chairman Russell has identified in his message transmitted in Kyndall Barry's E-mail message dated March 15, 2010.

**Chapter 6: Options for Elements of a Standard to Protect Against Effects from Aquatic Acidification**

**15. To what extent does the Panel agree that the proposal to develop an Atmospheric Acidification Potential Index (AAPI) linked to key determinants of aquatic ANC is a reasonable approach to developing an ecologically relevant standard? Does the Panel generally agree that the secondary standard options identified by staff (including indicator, averaging time, form, and level) are generally consistent with the available scientific and technical information and are appropriate for consideration by the Administrator?**

I believe that the structured plan outlined in Chapters 5 and 6 for an Atmospheric Acidification Potential Index (AAPI) linked to key determinants of aquatic ANC is a reasonable approach to developing an ecologically-relevant two-pollutant standard. A very complicated series of adjustments in thought processes by air quality managers will obviously have to be borne in mind. This will take a good deal of educational as well as technical thought.

But the logic and rationale behind the various parts of the AAPI as outlined in chapters 5 and 6 appear to be reasonably well thought out. Thus, I have so far discovered no serious omission from the various location-specific and ecosystem-specific factors as well as the mathematical calculations and modeling estimates that will have to be brought together when these kinds of integrated ecologically-relevant standards are implemented for two Criteria Pollutants at the same time.

Also, the attention given in Chapter 6 to each part of the definition of a NAAQS standard (indicator, level, statistical form, and averaging time) seem appropriate to me.

**16. What are the views of the Panel on the degree of uncertainty associated with each element of the suggested standard, e.g. the ecological indicator; the concentration to deposition ratios, reduced nitrogen, the natural background ANC, and the ambient indicator and averaging time for NO<sub>x</sub> and SO<sub>x</sub>, and its relationship to the degree of protection that could be expected from the standard? What are the views of the Panel on how to fairly characterize the uncertainty associated with the degree of protection that such a standard would provide from aquatic acidification?**

I am impressed with the extent to which the language used in Chapter 6 has embraced the concept of location-specific and ecosystem-specific Critical Loads which have been used widely in Europe and to a more limited extent in Canada. But finally, the USEPA seems to be coming

around to the idea that protection of ecosystems from adverse effects requires an approach that is very different from its traditional notions of national standards that are applied rather uniformly across our country for protection against adverse effects on human health.

Once again in Chapter 6, the issue of how to cope with the uncertainties associated with the degree of protection that any integrated NO<sub>x</sub>/SO<sub>x</sub> secondary standard would provide has not been dealt with very explicitly – except with regard to spatial variability. I hope that others in our panel, such as Rich Poirot, will have more to suggest than I have so far been able to suggest.

**17. What are the views of the Panel on aggregating the terms (Q, N<sub>eco</sub> and BC<sub>0\*</sub>) used in estimating natural background ANC, denoted by the function g(·), into 5 bins based on the geologic classification scheme?**

Use of a series of “5 (or more?) bins” for aggregating the terms (Q, N<sub>eco</sub> and BC<sub>0\*</sub>) used in estimating natural background ANC, denoted by the function g(·), seems reasonable to me, but I confess to having no direct experience from which to offer a well-informed professional judgment in response to this question.

**18. What are the views of the Panel on the use of regional air quality modeling (e.g., CMAQ) results to establish the concentration-to-deposition ratio (V<sub>NO<sub>y</sub></sub>, V<sub>SO<sub>x</sub></sub>) and reduced nitrogen deposition (NH<sub>x</sub>) in the AAPI calculation? What are the views of the Panel on the critical uncertainties associated with the use of CMAQ to generate these parameters, and the potential implications of these uncertainties for the secondary standards?**

Although I am not a very mathematical-modeling-savvy kind of scientist, I have come to have great respect for the usefulness of both the CMAQ and the MAGIC models, and thus offer a largely-second-hand assurance of professional confidence in the usefulness of these two specific models for atmospheric processes in the case of CMAQ and acidification effects on aquatic ecosystem in the case of MAGIC.

**19. What are the views of the Panel regarding presentation of the standards as a set of tradeoff curves for NO<sub>x</sub> and SO<sub>x</sub> associated with specific levels of AAPI, and specific values of the g, V<sub>NO<sub>x</sub></sub>, V<sub>SO<sub>x</sub></sub>, and NH<sub>x</sub> terms?**

Using a series of tradeoff curves for comparative evaluation of alternative NO<sub>x</sub> and SO<sub>x</sub> secondary standards associated with specific levels of AAPI, and specific values of the g, V<sub>NO<sub>x</sub></sub>, V<sub>SO<sub>x</sub></sub>, and NH<sub>x</sub> terms seems intuitively promising to me. But, once again, I must confess to having no direct professional experience from which to offer a well-informed professional judgment in response to this question.

**20. What are the views of the Panel on using a single year or a three year average of recent year CMAQ modeling results to estimate the AAPI terms?**

I agree with the suggestion in this draft Policy Assessment document that both one-year and three-year-long modeling results should be done for several different acidification sensitive and

potential nitrogen-enrichment sensitive geographical areas and compared in the next draft of this CASAC Policy Assessment NAAQS Secondary NO<sub>x</sub>/SO<sub>x</sub> Review Panel process.

**21. What are the views of the panel on the ambient monitoring requirements? Is for the proposed three ambient air measurements – NO<sub>y</sub>, SO<sub>2</sub> and particulate sulfate – sufficient to judge compliance with and AAPI?**

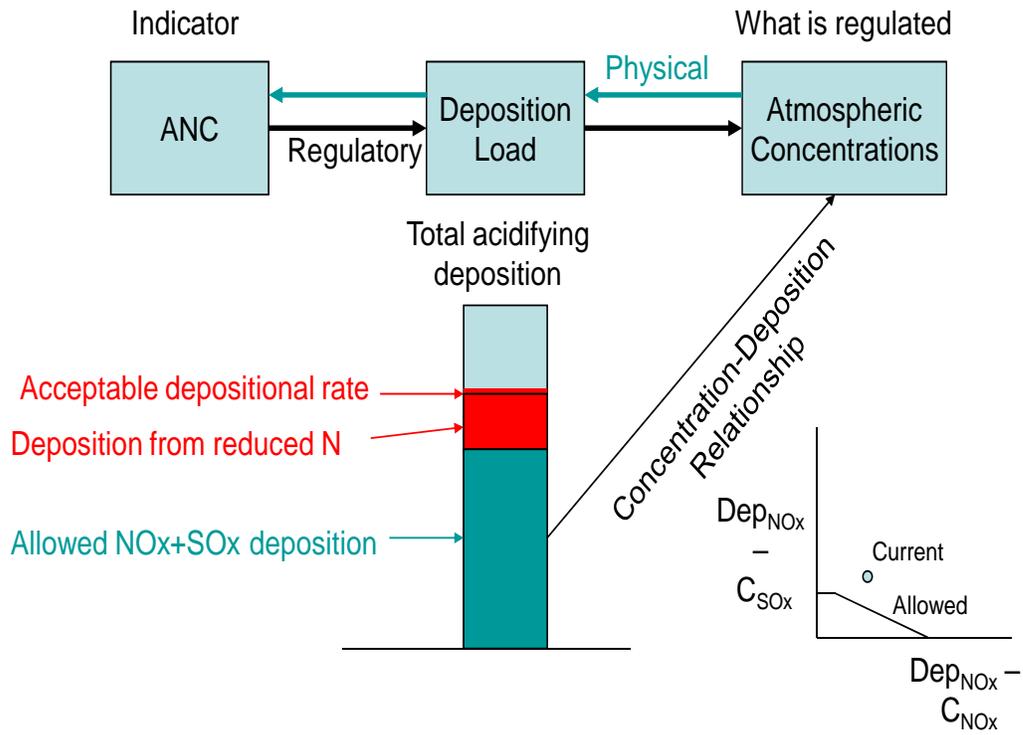
No, I believe these three ambient air measurements – NO<sub>y</sub>, SO<sub>2</sub> and particulate sulfate – would not be sufficient to judge compliance with an AAPI unless NH<sub>x</sub> is also included in the evaluation. It is the amount of total reactive nitrogen (NO<sub>y</sub> + NH<sub>x</sub>), and not just the chemically oxidized forms of nitrogen and sulfur that cause adverse effects on both terrestrial and aquatic ecosystems.

**22. What are the views of the Panel on using existing NO<sub>y</sub> and PM<sub>2.5</sub> sulfate measurement techniques as the basis for defining a Federal Reference Method to judge compliance with and standard?**

I believe that consideration should be given to using the existing measurements of NO<sub>y</sub> and PM<sub>2.5</sub> sulfate measurement techniques for defining a possible Federal Reference Method to judge compliance with a possible NO<sub>x</sub>/SO<sub>x</sub> Integrated Secondary Standard. But I also know that we will need a substantially increased number of remote, rural, suburban, and urban sites and must examine critically the frequency with which these techniques would be used and the results compiled in useful ways.

Below please find a simplified diagram developed by Ted Russell for a presentation he made to the Integrated Nitrogen Committee of the EPA Science Advisory Board in October 2009.

# Integrating Across Species Contribution to Acidification: A Proposed EPA Approach...



## **Dr. Charles T. Driscoll**

### **General Comments**

Modify the wording of the document by refraining from stating that inanimate objects like figures or chapters on documents state or do things.

Need to define acronyms that are not defined.

The referencing of the document is very uneven. Some references are provided. However in some sentences detailed information is provided without proper citation.

Please refrain from using the word reduce. The word reduce can mean to decrease, but it can also mean to chemically reduce. Confusion will be less if the word reduced was only used to indicate chemical reduction.

Also the word level is not a very specific term. Using it in the document leads to confusion. Does level refer to concentration or deposition? It is best to use the actual term of interest.

Deposition units should be kg (indicate mass basis)/ha-yr not kg/ha/yr.

P67. TMDLs. In addition to Chesapeake Bay, there are TMDLs for surface water pH in at least two states NY and TN. Shouldn't these be mentioned?

P157 (Section 5.2.2.2). The models used are steady-state models. I think some discussion of the advantages and disadvantages of steady-state vs. dynamic models is critical here. I think this would be an essential foundation to this and other sections. Without such a discussion, the section is lacking.

P164. I think it is a great idea to evaluate additional landscape features in categorizing landscape sensitivity. Elevation is a logical one. Others might include forest cover and watershed area.

P164, line 25 This statement is not entirely true and should be re-written. Reduced nitrogen can be converted to nitrate but not always. It can be assimilated into soil and biomass. In fact most studies show it largely goes into soil. It can be denitrified. Also I don't believe that nitrate deposition has the same effect as ammonium deposition. Generally it would not. It depends on the associated cation and anion respectively.

P167, line 20. I like the analysis of terrestrial and aquatic acidification. However, I suspect the analysis would be flawed by the application of steady-state models. Steady-state models assume that the soil exchange remains consistent over time. In reality, the base saturation of soil decreases with acidic deposition. So the sensitivity of terrestrial ecosystems to acidic deposition likely decreases with time. This aspect of acidification should be addressed in the text.

P178, equation 2. Is this equation (2) correct? It does not seem possible. Doesn't the lake retention term need to be prorated by its area? Does this equation assume some land cover other than forest and lake? Some description of this equation is essential to inform the reader of the intent behind it.

P193. This line of reasoning of short-term atmospheric events having important implications for episodic acidification is flawed. Episodic acidification largely results from shifts in hydrologic flow paths (Chen et al. 1984). Inputs of nitrogen and sulfur from snowpack and atmospheric deposition largely cycle through soil. Short-term direct inputs from atmospheric deposition are not important in episodic acidification.

P197, line 4 and elsewhere. I am concerned that the Neco term does not include organic nitrogen. This could be problematic. Some discussion of organic nitrogen should be given.

P214, line 2. Again, I question the validity of comparing terrestrial and aquatic ecosystem sensitivity given the steady-state modeling approach used. In some systems soil loss of exchangeable basic cations will continue over time. It would be more appropriate to use a dynamic model to evaluate this point.

### **Specific Comments**

P2, line 19	Change to: A summary of ecological... from the ISA and REA is provided in Chapter 2.
P2, line 21	Change to: Those ecological effects are placed within the context of "public welfare" in Chapter 3...
P2, line 23	Change to: The adequacy of the ... ecological effects is discussed in Chapter 4.
P2, line 24	Change to: The conceptual design ... multi-pollutant standards is developed in Chapter 5.
P2, line 25	Change to: Options for developing ... conceptual design are presented in Chapter 6.
P2, line 27	Change to: How secondary ... other ecological endpoints is described in Chapter 7.
P2, line 28	Change to: A consideration ... and oxidized forms of nitrogen is provided in Chapter 8.
P3, line 1	Change to: Preliminary staff conclusions are summarized in Chapter 9, regarding ...
P3, line 10	... NO <sub>x</sub> and/or SO <sub>x</sub> , but ...
P4, line 2	... including total reactive ...

P5, line 6	Change to: In the ISA the ecological ... to ecosystems are highlighted other ...
P5, line 7	Change to: In this assessment information on gas-phase ... on vegetation is evaluated, but effects of gas-phase NO <sub>x</sub> ... on multiple ecological receptors are emphasized.
P7, line 28	Change to: framework, in the next section, we provide ...
P8, line 5	draft Air Quality Criteria Document (AQCD) ...
P8, line 6	Change to: In this draft document a comprehensive assessment was provided ...
P12, line 1	Change to: In the petition other adverse ... criteria pollutants were also listed ...
P13, line 3	Change to: NO <sub>x</sub> and SO <sub>x</sub> for a combined secondary standard at this time
P13, line 6	Change to: The conceptual framework was introduced in the REA for ...
P13, line 14	Change to: The framework we are considering for the structure ... standard is depicted in Figure 1-1.
P13, line 24	What is meant by levels? Concentrations? Levels is not a very specific word. Please clarify.
P13, line 25	Change to: a particular extent of ecosystem protection are those concentrations that ...
P14, line 5	Change to: As more information is available on effects ... NO <sub>x</sub> and SO <sub>x</sub> , this approach ...
P14, line 9	Change to: such as atmospheric characteristics and ecosystem sensitivity.
P14, line 20	Change to: effects
P15, line 18	Change to: ... humans following inhalation of air pollutants ...
P15, line 26	Change to: ... discussed in the ISA, the REA and ...
P19, line 3	Change to: ... variables controlling the relationships of the ecological response to deposition.

P19, line 7	Change to: ... weathering rates, elevation, climatic factors or biological factors that ...
P19, line 9	What is meant by high natural background acidification? Do you mean high internal supply of naturally occurring organic acids? Please clarify.
P20, line 23	Change to: ... nitrogen deposition diminished when NO <sub>x</sub> related deposition is decreased?
P22, line 2	Change to: In this chapter we address ...
P22, line 17	Change to: Those effects are evaluated in Chapter 3 within the ...
P23, line 19	Change to: base saturation of soil decreases. Continued ... can deplete the available base cation pool in soil.
P23, line 22	Change to: The ability of a watershed to neutralize acidic deposition ... factors including weathering rates, bedrock composition, vegetation and microbial processes, physical and chemical characteristics of soil and hydrologic flowpaths (REA 2.1).
P24, line 1	Change to: increases in inputs of N and S to ecosystems and the associated ...
P24, line 2	These sentences are confusing. The first phrase refers to short-term deposition. The second part of the sentence and the following sentence refers to episodic acidification. I am confused. It appears that some text is missing.
P24, line 5	Change to: Episodic <u>acidification</u> refers ...
P24, line 20	Delete the last phrase in the sentence after the comma.
P24, line 24	Define surplus of base cations. Change to: ... in surface waters, the sum of base cations, ANC, dissolved inorganic aluminum and pH (Note that calcium is a base cation).
P25, lines 1 and 2	Change to: <u>dissolved</u> inorganic Al
P25, line 9	Change to: Similarly, <u>decreases in</u> prey ...
P25, line 19	Change to: about 15 to 20%, exchange
P25, line 21	Change to: <u>dissolved</u> inorganic Al. The effect ...
P25, line 28	Change to: <u>Acidic deposition and soil</u> acidification has ...
P26, line 3	Change to: <u>Sugar</u> maple

- P26, lines 10, 11, 19 & 20 Forests are the only terrestrial ecosystem type that is sensitive to acidic deposition. Should the word “terrestrial” be changed to “forest” in these sections and elsewhere to be more specific?
- P27, line 4 Change to: deposition in this region is less clear
- P27, line 23 Change to: integrity occur because the energy ...
- P28, line 12 Figure 2-1 Change to: Ecological effects associated with alternative levels of acid neutralizing capacity (ANC)
- P29, line 1 17 - and 5 - fold higher than what? Please clarify.
- P29, line 4 This is a mistake and needs to be corrected. Virtually all watersheds experience episodic acidification. Are you referring to acidic episodes (i.e., short-term decreases in ANC to low values that would cause biological effects)? Please clarify.
- P31, line 7 Change to: are less sensitive sites
- P31, line 8 Change to: Note that studies ...
- P31, line 15 Change to: commonly consumed by sport fish
- P31, line 7 Again as in the earlier Adirondack section 10 – and 32 – fold higher than what? Please clarify.
- P31, line 10 Again virtually all watersheds experience episodic acidification. Do you mean acidic episodes?
- P31, line 17 Change to: a decrease in the condition ...
- P 34, line 8 Change to: are less sensitive sites.
- P34, line 9 Would it be good to refer to “forest acidification” rather than “terrestrial acidification”?
- P34, para. starting at line 10 It is necessary to clarify the basis of the ratios. I believe these are molar ratios. If I am correct, they should be clarified as such (e.g., line 10 Bc/Al soil saturation molar ratios. Line 14 Critical soil Bc/Al molar ratio ... and throughout the remainder of the document.
- P35, line 16 Change to: ... can be decreased by 20% ... growth can be decreased ...
- P35, line 21 Change to: For red spruce
- P36, line 17 (i.e., ANC)
- P36, line 28 Change to: Terrestrial (Forest?) Acidification

P36, line 30 Change to: USFS – Kane Experimental Forest

P37, line 2 Change to: was decreased by 20%.

P37, line 4 Change to: by soil available Ca<sup>2+</sup> depletion ...

P37, line 21 Change to: nitrogen (e.g., NH<sub>x</sub>)

P38, line 8 Change to: chronic additions of N

P38, lines 13, 14, 15

P39, lines 7, 8

P41, line 2

P38, line 13 Change to: NO<sub>3</sub><sup>-</sup> leaching, soil C:N ratio, rates of N mineralization, nitrification, and denitrification ...

P38, line 29 & throughout doc. Units of flux should be kg/ha-yr, not kg/ha/yr.

P39, line 28 Change to: directly deposited to the water surface can pollute the surface water.

P41, line 15 Change to: Figure 2-8.

P42, Figure 2-8 Define MCF – Mixed Conifer Forest; CSS – Coastal Sage Scrub.

P43, Figure 2-9 Can these locations be reorganized so they are easier to follow, either west to east or east to west or terrestrial vs. aquatic. The numbers do not seem to appear in any logical order. Also change # 10 to elevated NO<sub>3</sub><sup>-</sup> leaching.

P45, line 29 Change to: NO<sub>3</sub><sup>-</sup>

P46, line 21 Change to: DO, decreases in biodiversity ...

P46, line 21 Change to: desired decrease in load

P46, line 23 Change to: must be controlled

P46, line 24 Change to: decrease of total nitrogen

P46, line 25 Change to: indicated decreases in atmospheric ... could not eliminate coastal ...

P46, line 27 Change to: by decreasing atmospheric

P49, line 5 Acronym correct? Define please.

P50, line 11 Change to: At sufficient concentration

P51, line 5 Change to: decreases in photosynthesis

P51, line 6 Change to: fixation, decreased k<sup>+</sup>

P63, line 9 Change to: to decrease or impair

P65, lines 4-8 This sentence is confusing. Can it be re-written?

P65, line 8 Change to: While the

P66, line 11 alkalinity is equivalent (not roughly, it is).

P67, line 27 There are also TMDLs for surface water pH. At least for the states of NY and TN. Shouldn't these be mentioned?

P72, line 1 delete etc. in paranthesis

P73, line 11 Change to: The spatial relationships ... deposition levels are illustrated in Figures 3-4 and 3-5.

P74, 75; Figures 3-4 and 3-5 Give the units of deposition in figures.

Figure 3-4 Change to: ... deposition of nitrogen and sulfur ...

Figure 3-5 Change to: ... deposition of nitrogen and sulfur ...

P77, line 21 Change to: (e.g. bald eagle – should be lower case)

P77, line 22 Change to: willingness to pay (WTP)

P78, line 16 In brief for each region experiencing ecological effects, ecological indicators ...

P78, line 22 The wording of this sentence is confusing. It should be re-written. Change to: figure

P79, line 4 Change to: In the next four sections, we summarize ...

P79, line 17 Change to: northeastern United States

P80, line 21 Change to: In the previous section, we describe the ecosystem ..., and summarize evidence ... existing NO<sub>x</sub>/SO<sub>x</sub> levels has not been quantified.

P81, line 9 Change to: In this case study, estimates of changes ... services are determined, as well ...

P81, line 15 What is meant by “zero-out” emissions? Please clarify.

P82, line 20 Bc:Al molar (?) to decreased tree ...

P82, line 27 Change to: northeastern United States

P83, lines 3, 10, 29 Change to: northeastern United States

P83, line 11 space after 2006

P85, lines 20, 28 decreases not reduction

P86, line 13 Change to: \$2 billion (add space between \$2 and billion)

P87, line 4 Change to: nitrogen decreases the ...

P88, line 30	Change to: nitrogen deposition <u></u>
P101, line 11	Change to: In this chapter, we address ...
P101, line 12	Change to: In the chapter, we begin
P102, line 9	Change to: as indicators, the current
P102, line 11	Should inputs of reduced N be mentioned here?
P102, line 16	Change to: acid inputs is <u>decreased</u> as natural buffers are <u>depleted</u> more rapidly than they ...
P103, line 11	Change to: In addition, in this chapter we qualitatively address ... SO <sub>x</sub> and set up arguments ...
P104, line 25	Change to: This information suggests...
P105, line 6	Change to: ... joint impact of the <u>multiple</u> pollutants ...
P105, line 8	Change to: This <u>information</u> suggests
P105, line 23-24	Change to: deposition falling on <u>base-rich</u> soils <u>underlying calcareous deposits</u> have a ... falling on shallow <u>acidic</u> soils ...
P105, line 25-26	Change to: detriment <u>to elevated acidic deposition</u> while ... may experience a <u>marked</u> loss in fish communities <u>to a similar input</u> .
P106, line 18	Change to: plus ammonium, NH <sub>4</sub>
P108, line 11	Change to: 4-8 sites in the <u>S</u> outheast ...
P110, line 9	Change to: and IMPROVE networks. In addition
P111, line 1	Change to: primary compliance purposes capture (delete “do”)
P111, line 4	<u>S</u> outheast, there are...
P111, line 22	Change to: of nitrate, ammonium, sulfate <u>and other</u>
P111, line 29	Change to: The <u>o</u> rganic nitrogen is ...
P112, line 8	Change to: deposition through <u>m</u> onitoring and <u>m</u> odels?
P113, line 19	define FAB
P114, line 114	define PAD
P115, line 10	Change to: reactive nitrogen and sulfur,
P115, line 25	Change to: throughout the <u>e</u> astern United States
P115, line 29	Change to: (NH <sub>3</sub> and NH <sub>4</sub> ),
P115, line 30	Change to: are provided in <u>f</u> igures 4-5
P116, line 3	Change to: In addition, <u>f</u> igures 4-12

P116, line12	Change to: associated with the <u>formation</u> of
P116, line 16	Change to: <u>Ambient</u> conditions include ...
P116, line 18	Change to: This <u>information</u> is consistent
P120, Figure 4-8	Change to: ammonia, NH <sub>4</sub>
P125, Figure 4-14	Change to: ... from reporting stations in <u>Air Quality System (AQS)</u> .
P126, Figure 4-15	Change to: ... CMAQ modeled oxidized nitrogen deposition (kgN/ha-yr).
P127, Figure 4-16	Change to: ... CMAQ modeled oxidized sulfur deposition (kgS/ha-yr).
P130, line 11	Change to: <u>In</u> the previous sections <u>we</u> have
P130, line 13	Change to: This <u>pattern</u> suggest that
P131, line 1	Change to: <u>In</u> this section <u>we</u> <u>focus</u> on ...
P131, line 7	Change to: (N deposition in the <u>eastern</u> U.S. includes ...
P131, line 8	Change to: locations with greater than 9 kg <u>N/ha-yr</u> ,
P131, line 12	Add reference
P131 and throughout	Change from ha/yr to ha-yr
P132, line 1	Change to: flux of methane (CH <sub>4</sub> ), and ...
P132, line 8	Change to: In Chapter 3 of the REA a thorough assessment is provided ...
P132, line 15	Change to: As discussed throughout the REA document, ...
P133, line 12	Change to: fully overlap. The locations of the case studies evaluated in the REA are shown on Figure 4-20.
P133, Figure 4-20	Change to: ...map highlighting the nine case study ...
P134, line 8	Change to: measuring the acid <u>neutralizing</u> capacity...
P134, line 16	Change to: and SO <sub>x</sub> and <u>loss of ANC in sensitive ecosystems</u> , and
P134, line 23	Change to: ... case study area is 62.1 <u>µeq/L</u> (moderate ...
P134, line 25	Change to: ANC of 50 <u>µeq/L</u> , and ...
P134, line 26	Change to: This <u>information</u> indicates ...
P134, line 28	Change to: ... with greatly <u>diminished</u> fish species ....
P135, line 10	Change to: 57.9 <u>µeq/L</u> , indicating ...

P135, line 12	Change to: ... area, this <u>information</u> suggests ...
P135, line 18	Change to: In the ISA it is noted that large portions of the eastern U.S. ...
P135, line 21	Should also note that the deposition is much lower.
P135, line 26	A reference should be provided for this report.
P136, line 1	Change to: ... could be classified as <u>acid-impacted</u> based on ...
P136, line 2	Change to: ... lakes classified as <u>acid-impacted</u> based on chronic
P136, line 5	Change to: ... in the <u>eastern</u> U.S. ...
P136, line 9	Change to: ... services such as recreational fishing.
P136, line 24	Change to: ... with significant <u>effects on</u> ...
P137, line 1	Change to: ... sugar maple growth can be <u>decreased</u> by 20 percent.
P137, line 10	Change to: ... for a Bc/ <u>Al</u> ratio ...
P137, line 13	Change to: ... range for sugar maple (e.g. Arkansas, Illinois) no plots ...
P137, line 16	Change to: ... loads for a BC/ <u>Al</u> ratio of ...
P137, line 18 and 19	Need a reference
P137, line 22	Do you mean concentrations rather than levels?
P138, line 23 and 24	Change to: 150 kg/ha-yr and 300 kg/ha-yr .... 0 to 5.5 kg/ha-yr
P138, line 30	Change to: In the ISA it is noted that ...
P138 & 139, line 31 & 1	Change to: In addition, forest management practices can significantly affect the nitrogen cycling, and ...
P139, line 17	Change to: in those locations <u>decreases</u> ...
P139, line 30	Change to: ... deposition <u>are</u> limited,
P140-141	Change all ha/yr to ha-yr
P142, line 26	Many watersheds throughout North America may be conducive to methyl mercury production (e.g. Everglades, Southeast).
P145, line 11	Change to: There are now sufficient data ...
P145, line 14	Change to: (e.g. elevation, groundcover)
P145, line 19	Change to: ... are presented in <u>Chapter</u> 6.
P146, Figure 5-1	Change to: ... conceptual design of the NO <sub>x</sub> and SO <sub>x</sub> secondary standard.
P146, line 20	Change to: In the following section, we describe ...

P147, line 9	Change to: ... NH <sub>x</sub> is often a <u>large component</u> of the ...
P147, line 13	Change to: ... application to aquatic acidification <u>are</u> presented ...
P147, line 20	Need to define ANC limit.
P147, line 23	Change to: ... is known, <u>one</u> could calculate ...
P148, line 17	Define tradeoff curve.
P149, line 11	Change to: In this section we present the ecological components ...
P149, line 20	Change to: ... acidification <u>is</u> one that is measurable
P149, line 26	Change to: ... indicators that <u>could</u> be used ...
P150, line 15	Change to: ... input of <u>strong</u> acid anions (e.g. NO <sub>3</sub> <sup>-</sup> , SO <sub>4</sub> <sup>2-</sup> ).
P150, line 16	Change to: ... indirectly via <u>drainage from</u> terrestrial ecosystems. ... when the <u>strong acid</u> anions are ...
P151, line 1	Define F-factor
P151, line 3 & 4	Change to: ... N and S deposition on <u>predicted pre-industrial ANC value</u> . Note that acidification ...
P151, line 15	Change to: ... sulfur deposition, <u>although this relationship is altered by landscape factors (e.g. geology, soils, land cover)</u> .
P151, line 22	Change to: ... two case study areas. <u>These</u> data <u>were</u> used to compare ...
P151, line 23	Change to: ... (i.e. preacidification, 1860).
P152, line 1	Change to: ... <u>marked</u> increase in the number of <u>acid-impacted</u> lakes, characterized ...
P152, line 14	Change to: ... correspond to important <u>conditions</u> along the ANC response <u>surface</u> that are ...
P152, line 19	Define critical limit of deposition.
P152, line 23	Change to: ... (REA 4.2). Note that a <u>given</u> level of ANC ...
P152, line 25	Change to: ... concludes that a <u>quantitative</u> relationship ...
P153, line 8	Change to: that will result in adequate biogeochemical conditions to sustain ecosystem health. Adequate biogeochemical conditions is a subjective term ... (e.g. ANC = 20, 50, 100 µeq/L) representing different degrees of protection of aquatic ecosystems against acidic deposition.
P153, line 13	Change to: base cation <u>supply</u> to an ecosystem
P153, line 14	Change to: response to <u>acidic</u> deposition

P153, line20	Change to: models <u>are</u> rather extensive
P154, line 3	Change to: modify the <u>surface water</u> ANC
P154, line 25	Change to: <u>A</u> ppendix 4
P154, line 27	Why the New England EMAP probability survey and not the Adirondack EMAP survey?
P155, line 12	Change to: include;
P155, line 13	Change to: model;
P155, line 14	Change to: concern;
P155, line 25	Change to: data <u>are</u>
P156, line 2	Change to: parameters <u>are</u>
P156, line 4	Change to: that <u>are</u>
P156, line5	Change to: scale <u>are</u> not
P156, line 26	Change to: include; (1) ... model; (2)
P156, line 27	Change to: concern;
P156, line 29	Change to: chemistry <u>are</u> not
P157, line 17	Change to: <u>steady-state</u> SSWC
P157, line 21	font shift ????
P157, line 24	Change to: is incorrect <u>because</u> in many <u>ecosystems</u> nitrate
P157, line 25	Change to: If <u>nitrate</u> is leaching
P157, line 25	“Nitrate leaching is determined from the sum of measured concentrations of nitrate and <u>ammonium in the runoff</u> ”. This sentence makes no sense. Admittedly in many systems concentrations of nitrate are low, but really. What about organic nitrogen? Why is this ignored? It should be at least addressed in the text.
P158, line 1	Change to: calculation <u>are</u>
P158, line 9	Change to: ; 2
P158, line 10	Change to: ; and 3)
P158, line 16	This figure title is not very detailed or descriptive. A more detailed figure title should be given.
P159, line 3	Change to: µeq/L
P159, line 4	Change to: µeq/L

P160, line 15	Change to: model <u>are less certain</u> .
P160, line 26	Change to: indication <u>to be utilized is</u> ANC.
P160, line 28	Change to: is known, the
P161, line 2	I Change to: <u>In</u> the following discussion, <u>we</u>
P161, line 4	Change to: loads <u>is</u>
P162, line 12	Change to: (Figure 5-4)
P162, line 13	Change to: Sullivan et al. (2007)
P162, line 17	Change to: Sullivan et al. (2007)
P163, line 9	Change to: <u>have greater soil depth</u> receive
P164, line 25	This statement is not entirely true and should be re-written. Reduced nitrogen can be converted to nitrate but not always. It can be assimilated into soil and biomass. In fact most studies show it largely goes into soil. It can be denitrified. Also I don't believe that nitrate deposition has the same effect as ammonium deposition. Generally it would not. It depends on the associated cation and anion respectively.
P165, line 21	Change to: <u>from</u>
P170, line 20	Change to: <u>A</u> relationship for ... concentrations is provided in Equation 5.
P171, line 2	Change to: aggregated deposition (space)
P172, line 25	Change to: <u>The</u> oxidized sulfur ... values are listed in Table 1.
P173, line 1	Change to: simulations of (space)
P173, line 5	Change to: <u>Annual</u> inverse ... simulations <u>are shown</u> in Figure 5-5.
P173, line 8	Change to: SO <sub>x</sub> <u>concentrations</u>
P173, line 9	Change to: <u>An</u> example ... ratios <u>is</u> shown in Figure 5-6.
P173, line 10	Change to: Section 6 for (space)
P173, lines 11, 15	Are these figure numbers correct? Figure 1, Figure 3
P173, line 15	Change to: CV values ... areas (Figure 5-7)
P174, Figure 5-5	It is difficult to read the scale of this figure.
P175, line 2	Change to: schematic <u>diagram</u>
P177, line 3	Change to: A proposed ... application is described in Section 5.2.
P177, line 5	Change to: <u>strong</u> acid anions
P177, line 9	Change to: <u>aquatic</u> biota,

P177, line 12	Change to: <u>strong</u> acid anions
P177, line 16	Change to: <u>In</u> this section, <u>we</u> summarize and provide ... the approach to calculate: (1) ... level; (2) ... level; and (3) ...
P178, line 3	Change to: <u>μeq/L</u>
P178, line 4	Change to: <u>μeq/L</u>
P178, line 25	Change to: <u>can</u> contribute to acidification
P178, line 28	( <u>NO<sub>x</sub></u> + S)
P179, line 12	Change to: <u>evaluated</u>
P179, lines 18, 19	50 <u>μeq/L</u>
P180, Table 5-4	Change to: Example <u>calculations</u> from <u>determining</u> the <u>percent</u> of <u>water</u> <u>bodies</u> <u>achieving</u> <u>target</u> ANC <u>levels</u> ... and <u>conduct</u> analysis right hand column ANC ≥ 50 <u>μeq/L</u>
P180, line 2	Change to: 50 <u>μeq/L</u>
P182, line 9	Change to: 50 <u>μeq/L</u>
P182, line 11	Change to: Neco is eliminated
P182, Table 5-6	Change to: 50 <u>μeq/L</u> ... units are in meq/m <sup>L</sup> -yr
P183, Figure 5-10	Change to: <u>NO<sub>x</sub></u>
P184, line 1	Change to: m <sup>2</sup>
P184, Figure 5-12	Change to: <u>NO<sub>x</sub></u>
P185, Figure 5-13	Change to: <u>NO<sub>x</sub></u>
P186, line 1	Change to: that <u>neutralize</u> the acidifying
P186, line 5	Change to: will vary in <u>Equations</u> 1 and 5
P186, line 19	Change to: is <u>separated</u> into
P190, line 3	What is by “mixture of the criteria pollutant(s). Please clarify.
P190, line 4	Change to: whether an area <u>is in attainment of</u> the
P190, line 7	Change to: (e.g. annual average).
P190, line 7	The term level is not clear. Please clarify. Do you mean concentration?
P190, line 8	Change to: specific <u>concentration</u> (?) to ...
P190, line 22	Change to: <u>A</u> conceptual framework ... standard <u>was described in Chapter 5</u> .

P190, line 27	Change to: Chapter 5, a set of potential ...
P191, line 13	Change to: <u>In</u> Section 6.2, <u>we</u> discuss
P191, line 14	Change to: <u>In</u> Section 6.3, <u>we</u> suggest
P191, line 15	Change to: <u>In</u> Section 6.4, <u>we</u> provide
P191, line 17	Change to: <u>In</u> Section 6.5, <u>we</u> discuss
P191, line 18	Change to: <u>In</u> Section 6.6, <u>we</u> address
P191, line 19	Change to: <u>Finally in</u> Section 6.7, <u>we</u> conclude
P192, line 9	Change to: S <sub>2</sub> O
P192, line 25	Define IMPROVE, if not previously defined.
P193, line 7	Change to: <u>Note</u> that chemical
P193	This line of reasoning of short-term atmospheric events having important implications for episodic acidification is flawed. Episodic acidification largely results from shifts in hydrologic flow paths (Chen et al. 1984). Inputs of nitrogen and sulfur from snowpack and atmospheric deposition largely cycle through soil. Short-term direct inputs are not important in episodic acidification.
P194, line 17	Change to: (See 75 FR 2938, 2999; January 19, 2010)
P195, line 11	Change to: ecosystem, <u>can potentially consume the acid neutralizing capacity</u> of the ... to additional loading from <u>acidic</u> deposition.
P195, line 20	Change to: to <u>re-evaluate</u>
P196, line 6	Change to: are <u>atmospheric</u> concentrations
P196, line 12	Change to: <u>Equation</u> (1)
P196, line 18	Change to: As such, <u>the</u>
P197, line 4 and elsewhere	I am concerned <u>that the Neco term does not</u> include organic nitrogen. This could be problematic.
P197, line 7	Change to: <u>Chapter</u> 4
P197, line 23	Change to: <u>variable</u>
P197, line 28	Change to: <u>measured</u>
P198, line 1	Change to: (e.g., to develop ... and N <sub>x</sub> )
P198, line 14	Change to: <u>sensitive</u>
P198, line 19	Change to: <u>Note</u> for this form
P198, line 20	Change to: different combinations (space)

P198, lines 21, 22	Change to: <u>E</u> quation (1)
P198, line 23	Do you mean g?
P200, lines 7, 8, 10, 12	total nitrogen retention (?) The term nitrogen buffering doesn't make much sense. Do you mean something else like retention?
P200, line 13	Change to: to <u>watershed nitrate leaching and associated acidification</u> .
P200, line 16	Change to: 50 <u>μeq/L</u>
P200, lines 18, 22	Do you mean watershed rather than water body?
P200, line 25	Do you mean g?
P201, line 2	Change to: areas (e.g. sensitive... Figure 6-1; reproducing ...
P201, line 9	Change to: (e.g. the 75 <sup>th</sup> or 95 <sup>th</sup> ... aquatic ecosystems),
P201, line 9	Do you mean g?
P201, line 16	Change to: from <u>decreases</u>
P201, line 25	Change to: naturally <u>acidic</u>
P201, line 26	Change to: naturally <u>acidic</u> areas, <u>decreasing</u> deposition
P203, line 21	Change to: <u>The</u> rational ... is discussed in Chapter 5
P204, line 2	Change to: natural <u>acid neutralizing</u> capacity
P204, line 24	Change to: also <u>reaches</u> a maximum at ... This <u>pattern</u> suggests
P205, lines 1, 17, 18, 23, 24, 25, 26, 28, 30	Give unit 50 <u>μeq/L</u>
P205, line 4	Change to: lead to <u>decreases</u>
P205, line 17	Change to: show a <u>marked</u>
P205, line 18	Change to: This <u>pattern</u>
P205, line 20	Change to: to <u>decrease</u> the
P206, lines 1,2	Change to: 100 <u>μeq/L</u> (give units)
P207, lines 9, 10, 19, 20	Change to: 50 <u>μeq/L</u> (give units)
P208, line 7	Change to: (Section 3.2)
P208, line 12	What is NCore? Please clarify.
P208, line 15	Define EPA CSN, if not done previously.
P209, line 28	Change to: largely oriented <u>towards populated areas</u>
P210, line 1	Change to: every watershed <u>would not be feasible</u> and may not ...
P210, line 11	Change to: the <u>NO<sub>x</sub></u> and <u>SO<sub>x</sub></u> <u>Secondary</u> <u>Standard</u>

P210, line 27	Change to: (e.g. ANC may be <u>naturally low</u> )
P210, line 9	Change to: assessment <u>l</u>
P211, lines 10, 24, 26, 28	<u>µeq/L</u> (give units)
P211, line 20	Change to: large <u>decreases</u>
P212, line 5	Change to: 100 <u>µeq/L</u>
P213, line 4	Change to: <u>In</u> this chapter, <u>we</u> focus
P213, line 11	Change to: NO <sub>x</sub> <u>and</u> SO <sub>x</sub> <u>Secondary</u> Standard
P213, line 14	Change to: 50 <u>µeq/L</u> , <u>soil</u> Bc:Al <u>molar</u> ratio
P213, line 21	Change to: 50 <u>µeq/L</u>
P213, lines 22, 23, 24, 26	Change to: <u>soil</u> Bc:Al <u>molar</u> ratio
P213, line 26	Change to: <u>Table</u> 7.1
P214, Table 7.1	Change to: ANC 50 <u>µeq/L</u> ... soil Bc:Al molar ratios ... lakes or streams
P214, line 2	Again I question the validity of comparing terrestrial and aquatic ecosystem sensitivity given the approach used. In some systems soil loss of exchangeable basic cations will continue over time. It would be more appropriate to use a dynamic model to evaluate this point.
P214, line 11	Change to: <u>soil</u> Bc:Al
P217, line 31	Change to: aquatic <u>ecosystems</u> ;
P218, line 2	Change to: impacts;
P219, line 18	Change to: in the <u>REA</u>
P220, line 19	Change to: by <u>decreases</u> in ANC
P220, line 22	Change to: 50 <u>µeq/L</u>
P221, lines 4, 7, 10, 14, 15	Change to: <u>soil</u> Bc:Al
P221, line 22	Change to: SO <sub>x</sub> ;
P222, line 9	Change to: S <sub>2</sub> O
P222, line 24	Change to: standard (e.g. the target ANC)
P224, line 1	Change to: <u>NO<sub>x</sub> and SO<sub>x</sub> Secondary</u> Standard

**References:**

Chen, C. W., S. A. Gherini, N. E. Peters, P. S. Murdoch, R. M. Newton, and R. A. Goldstein.  
1984. Hydrological analysis of acidic and alkaline lakes. *Water Resources Research*  
**20**:1875-1882.

## **Dr. Paul J. Hanson**

### **General Comment:**

The Policy Assessment for NO<sub>x</sub> and SO<sub>x</sub> welfare effects represents a good beginning. I agree with EPA staff that a useful NAAQS for NO<sub>y</sub> and SO<sub>x</sub> welfare effects requires a unique indicator, averaging time; form and level separate from the primary standards for NO<sub>x</sub> and SO<sub>x</sub>. I also agree that the data for terrestrial effects are not as well established as those for aquatic impacts. The proposed focus on the development of a standard focusing on aquatic considerations is an appropriate path forward.

The following comments are provided for EPA staff's consideration in the preparation of a second draft Policy Assessment (PA). I'm commenting on what I didn't like or what I would like EPA to change. In general, I found the document to be an appropriate discussion of the policy needs for the NAAQS process for secondary effects of NO<sub>x</sub> and SO<sub>x</sub>.

### **Front Matter – Key Terms**

The term or concept of “Adverse Effects” needs an overarching definition. I realize that Chapter 3 is set aside to discuss this issue, but a brief introduction to someone that might not read the entire report is advised.

In the definition of “Acid Neutralizing Capacity” I wondered if the word “water” shouldn't be replaced by precipitation. Pure water is not what falls from the sky.

Is base saturation really a measure of soil acidification (a process), or just an indication of the state of acidity of a given soil?

Add a definition of “Reduced N” to the list of key terms.

Should the definition of Eutrophication highlight its special significance to aquatic systems?

The definition of Semi-arid regions still contains a lower level of precipitation than the definition of arid regions. I've pointed this out previously and it hasn't been addressed. Someone needs to explain to me why the desert isn't as dry as the semi-desert.

Page xiii line 28: Super and subscripts need attention.

### **Chapter 1:**

Page 5 lines 6 to 11: Something isn't quite right in this section. The wording implies a heavy emphasis on direct effects of gaseous forms of NO<sub>x</sub> and SO<sub>x</sub>, but I was left with the impression that the intention might have been something else.

Page 12 lines 2 and 3: The inclusion of global warming in this list seems unnecessary and perhaps a bit of a stretch. Connections between NO<sub>x</sub> and SO<sub>x</sub> and climate change do not show up elsewhere in the PA document.

Page 13 line 26: Isn't the concept of "excessive degradation" a policy rather than a science question. Levels of change that represent excessive degradation are a judgment not a fact.

Page 14 line 9: The term "ecosystem sensitivities" might be replaced by 'location-specific characteristics' to better inform the new reader of the intent.

Page 17 lines 12 and 13: I don't think the PA does a very good job of addressing this question.

Page 20: On this page and in some other locations within the document the concept of reduced N (meaning chemically-reduced forms of N) may be confused with discussions of reduced deposition of N forms (see lines 23 and 24). I know what the authors intended, but I did stumble over the presentation.

Page 21 line 18: Does the Agency or the Administrator alone actively consider the appropriate levels?

## **Chapter 2:**

Page 23 line 8: Deposition of acidic compounds does lead to exposures, but not necessarily effects. The relationship between deposition (the exposure) and the effects depends on the characteristics of the ecosystem.

Page 23 line 11: The wording might be changed to "magnitude and rate".

Page 23 line 26: The phrase "small spatial scales" is not defined. Do the authors mean meters, kilometers, watersheds, counties, states or something else?

A common critique that I have within the PA document is that sentences are often written to suggest that acidification or nutrient enrichment effects have the potential to effect all ecosystems, vegetation or aquatic systems the same. The authors should be careful to reemphasize that many effects being discussed are applicable only to the most sensitive components of ecosystems. They may still warrant special protection, but they do not represent a response that is universal for all organisms.

Page 24 lines 21 and 22: Later on in the document confounding effects are mentioned, but this statement makes it appear that the results of acidification are absolute. In many cases I think it is likely that the evidence is based on inference drawn from strong correlations rather than a controlled study designed to isolate the specific effects of NO<sub>x</sub> and SO<sub>x</sub> additions to natural systems.

Page 25 line 17: Should the C: N ratio really be in this list. It is not emphasized in the document.

Top of page 26: The statements in this section do not suggest lots of confidence in the certainty of the relationship between acidification and observed phenomenon. The text describes a correlation, a response that “may be contributing” and a “likely” response. If appropriate, please change this text to indicate what we do know versus what continues to be hypothesized pending better data.

Page 26 line10: Not all terrestrial ecosystems are sensitive. The wording needs to be changed.

Throughout the document I believe it is important to characterize all pre-acidification results derived from MAGIC or CMAQ as simulations. Don’t pretend that we know what pre-acidification levels really are.

Page 32 line 6: I would change the wording here to “above simulated pre-acidification (1860) conditions”.

Page 38 line 4: The wording “terrestrial ecosystems” should be “sensitive terrestrial ecosystems”.

Page 38 lines 25 and 26: This is not true for all species, organisms, or ecosystems. Make sure that the reader is reminded that this document is taking the approach of protecting sensitive organisms and ecosystems.

Page 39 line 20: Add a reference for this statement.

Page 39 line 22 and line 24: The phrases “may result” and “may lead to” do not make a believer out of me. I would hope that we would be making suggestions for NAAQS based on data that can be fully supported by the published literature.

Page 40 lines 6 to 8: This statement is too strong. I don’t believe that “almost all” ecosystems will be altered by the addition of anthropogenic nitrogen. I actually expect the opposite. In most ecosystems (judged by % area of the US) we will be unable to detect the influence anthropogenic N additions. This is another case where the focus on sensitive ecosystems needs emphasis.

Page 40 line 13: I would add plant growth rate to the list of factors.

Page 43 line 11: “Hardwood forests” probably needs to be changed to sugar maple species. I doubt seriously that the vast majority of upland oak hardwood forests located throughout the eastern United States are very sensitive.

Figure 2-8 is pretty, but its application to the analysis isn’t obvious.

Within Figure 2-9: Are the conclusions cited under #13 the result of natural deposition or those derived from the manipulation studies conducted on the Fernow Experimental Forest?

Section 2.3.3: I was surprised that this section did not conclude with the strong statement from the ISA report that few direct effects from gaseous NO<sub>x</sub> or SO<sub>x</sub> are anticipated throughout most of the US.

Page 52 line 7: What does “significant numbers” imply?

### **Chapter 3:**

General comment: The PA document doesn't address natural rates of change versus change induced by anthropogenic N and S inputs. Change happens anyway. How are natural rates of change viewed in the context of other changes driven by N and S inputs or various other confounding factors?

Page 68 lines 23 to 29: I found this section of text to be very important. The realization that the establishment of critical load demands a judgment call of what is or is not an important adverse effect makes subsequent discussion of the use of critical loads in the characterization of risk to be a bit of a circular argument. This is an interesting conundrum. Does the EPA staff have a solution?

Page 72 lines 10 and 11: What is the policy relevant background level of ecosystem change or biodiversity alterations against which anthropogenic N and S induced effects might be judged?

Page 83 line 2: Provide the reference for this statistic.

Page 84 lines 7 to 23: I don't believe this paragraph is a strong argument for broad national controls. The available data are representative of only a few key species. Furthermore, if the net annual effect is only \$690,000 thousand dollars of production per year one might ask why we are concerned – certainly not for financial reasons. Conducting expensive research to understand responses, or setting standards to combat a problem that only sums to a \$690K per year phenomenon are not good justifications. The arguments based on endangered species are stronger.

Page 88 lines 9 to 15: These statements do not seem to consider that N and S deposition effects are fractional. For example, they are probably not likely to remove the entire ecosystem service for recreation. How does EPA judge the impact on aesthetic use if only a portion of an ecosystem is impacted by N and S inputs?

### **Chapter 4:**

Page 103 lines 3 and 4: I think this sentence should emphasize sensitive ecosystems not all ecosystems.

Page 131 line 28: Add the word ‘sensitive’ to qualify terrestrial and aquatic in this statement.

Page 131 line 31: Should carbon really be in this list? Convince me.

Page 132 lines 1 to 5: These items seem to come out-of-the-blue. Are they needed?

Page 134 line 21: Who defines the benchmark levels?

Page 135 line 18: The phrase “large portions” needs to be quantified. The authors should provide the area of the US involved to convince me that this is a big deal.

Page 136 line 16: There is not a significant risk to all terrestrial ecosystems. Only the sensitive systems are being highlighted in the case study analyses.

Page 136 line 28: Provide a quantitative statement for the land cover that defines “many” in this sentence.

Page 137 line 19 and 20: What criteria were used to define the critical load that forms the basis for this conclusion? Change the word “believes” to something else if you can. I’m not interested in what EPA staff believes. I want to know what they can justify from the science. Please conduct a search for the word believe and make appropriate substitutes throughout the document.

Page 138 lines 30 and 31: This may be true for some ecosystems, but other forests sequester all of the annual N inputs into aboveground wood production.

Page 139 line 8: Please be specific about the tree species to which this conclusion applies. It is not true for all species.

Section 4.5.2 seems to be missing a strong conclusion statement.

Section 4.5.4 seems a bit at odds with the statements in Section 4.5.2.

Page 141 line 5: “Large areas” is too vague. Please provide a quantitative statement about the amount of land area impacted.

## **Chapter 5:**

Page 145 line 14: You might add wind to the list of factors.

Section 5.3.1: This section reads as if the relationships are universal to all tree species. I don’t believe that EPA would be able to support that concept. Make this discussion reflect the sensitive species that it targets.

Section 5.4.1: In an attempt to make the equations simple and easy to follow the authors have not appropriately reflected what is really being attempted. Equations 1 through 4 should include components for gaseous, particle and occult deposition that depend on concentrations and deposition velocities plus an independent component of wet deposition that is a function of rainfall. The processes are not the same. Please make the equations reflect the true processes being calculated within the model (e.g., page 172 lines 8 to 11).

**Chapter 6:**

No comment at this time.

**Chapter 7:**

This section is underdeveloped. There is not much to comment on.

**Chapter 9:**

Throughout this section make sure that the focus on sensitive aquatic and terrestrial ecosystems comes through loud and clear. Don't lead the reader to conclude that all ecosystems are uniformly at risk from N and S inputs.

In the research needs list a better precipitation model or emphasis on direct measures of wet deposition inputs should be further emphasized.

## **Dr. Dale W. Johnson**

### Comments on the Executive Summary:

P. ES-3, paragraph 5: I do not agree that geology is the principal factor governing sensitivity to N: decades of biogeochemical research have shown that plant uptake is a major factor (often THE major factor) deciding whether atmospherically-deposited N passes through the terrestrial ecosystem to the aquatic one.

p. ES-4, paragraph 3: Again, as the authors are so often loathe to admit, I would assert that N deposition to N-limited commercial forests could be beneficial. After many such comments, however, I do not, expect to see this fact reflected in any executive summary and this is unfortunate because I can assure you that my colleagues, at least, will take note of that and possibly dismiss this entire endeavor.

p. ES-5, last paragraph: The statement about the Sierra Nevada is an over generalization – the eastern and more remote portions of that range do not receive N deposition levels above 5.

p. ES-7, last paragraph: The statement that reduced forms of nitrogen are converted to nitrate is incorrect as a blanket statement: ammonium can be taken up by plants or microbes before being nitrified and, in N-limited ecosystems (that is, in 90% of all terrestrial ecosystems), this is the rule not the exception. Also, in the same paragraph, I simply do not understand the wording of the last sentence.

p. ES-8, paragraph 1: The treatment of  $\text{NH}_4$  is very cursory and no doubt will invoke much criticism.

p. ES-8, last paragraph: You should mention that this applies to aquatic ecosystems – there is no measure of ANC in terrestrial ecosystems as such.

p. ES-9, paragraph 2: give units. Also, what kind of term converts deposition to fluxes? I thought deposition WAS a flux?

p. ES-11, paragraph 1: Give units. Is something missing in the equation as per “( )”?

### Comments on the overall document:

I reviewed the entire document, with special attention to those portions where I have some expertise. Specific suggestions and comments are given below.

p. 23, line 9: replace ”in some instances” with “unless buffered by high base soils”

p. 23, lines 15-28: Good summary, but you should also mention the effects of  $\text{NH}_4^+$

p. 24, line 16: replace “leads to the acidification of” with “leads to varying degrees of acidification” . It is all a question of amount: small background levels of SO<sub>x</sub>, NO<sub>x</sub> and NH<sub>x</sub> will not lead to any significant acidification nor will this take place in limestone bedrock systems. As a generic statement, this does not stand.

p. 24, line 24: what is a surplus of base cations?

p. 34, line 12: Replace “Tree species” with “Some tree species”. Some tree species tolerate Al very well and in fact seem to thrive in acidic environments.

p. 35, line 4: Replace “exceeds” with “exceed”

p. 37, lines 8-12: Having worked with many models of acidification over the years, I definitely do not share your high confidence in this one. I am not sure what change is needed, but I want to go on the record as a skeptic.

p. 39, line 24: insert “deemed desirable in commercial forests but may also” in front of “may”. Bias toward negative is showing through here again and I believe it is important to express both sides of the nitrogen issue.

p. 70, Table 3-1: Where are forest products? Fiber, timber, furniture wood?

p. 72, lines 1-15: Important ecosystem services from forests should certainly include timber, furniture wood, pulp stock from commercial forests.

p. 82, lines 27-28: A very true statement – but where is this shown in Table 3-1?

p. 84, line 11: What about loblolly pine? It is far more commercially important than sugar maple or red spruce.

p. 84, lines 21-23: This is a gross overstatement: these estimates are based on two species and do not include the far more commercially important southern pines!

p. 84, lines 25-29: Why on earth have you decided to overlook any beneficial effects in your economic assessment? I cannot imagine this being defensible and certainly do not agree with it in any way.

p. 88, line 30: change “depositio” to “deposition”

p. 89, lines 1-2: If you select two case studies based on the presumed negative effects of atmospheric deposition, how can this possibly be any kind of regional assessment?

p. 102, line 11: Add “or NH<sub>4</sub><sup>+</sup>” after “SO<sub>x</sub>”

p. 136, line 20: BC:Al in soils, presumably?

p. 136, line 26: first mention I see of timber – again, loblolly pine is king of that.

p. 137, lines 1-8: This could use some references cited.

p. 138, lines 10-15: This section is biased. Nitrogen limitation is common, not only in limited situations and N inputs will probably cause long-term benefits in most managed systems. I recognize from past experience that the authors are very loathe to admit this, so I will continue to point it out. Your statements fly in the face of many decades of forest nutrition research.

p.138, lines 30-31 and on to page 139: As I have noted in the past, the statement that most atmospherically deposited N is retained by soils is disputed. The sentences after this try to minimize N benefits to commercial forests, as the authors have done in the past. We have agreed to disagree on this matter, it seems.

p. 153, lines 7-18: This is where I have real trouble with the model used: no ecosystem or soil is ever in steady-state; Were this the case, no soils would ever acidify in the absence of atmospheric acid inputs, and they certainly have done so as evidence by many studies in pristine areas such as Alaska.

p. 155, lines 18-20: This seems like an overly confident statement – what is there to back it up? For example, here in the Sierra Nevada, we have chronically hydrophobic soils which can profoundly affect surface hydrology in ways very different from eastern soils which are not hydrophobic. This is a gross over generalization.

p. 158, line 21 through p. 159 line 17: Please provide units for  $DL_{ANClim}(N+S)$  – it would seem from the other units given that the units for this would be  $meq\ m^{-2}$  after some conversion?

p. 177, line 26 through p. 178, line 11: Same comment

#### Comments on Chapter 7 Charge Questions:

### **23. What are the views of the panel on the approach taken to compare the protection provided by a potential aquatic acidification standard to the protection needed for terrestrial acidification?**

While I do not share the authors' faith in the validity of the models used, I see no other viable approach for answering this question. Thus, I can offer no criticism for the approach taken.

### **24. What are the views of the panel on a future comparison of the protection provided by a potential aquatic acidification standard to nutrient enrichment benchmarks? What are the views of the panel regarding using a nutrient enrichment benchmark to be a limiting factor on the nitrogen in the aquatic acidification standard, instead of having a separate standard?**

Acidification and nutrient enrichment could be very much decoupled, especially if the major cause of acidification is S and not N. Also, there will be little aquatic effect of N deposition until

the terrestrial ecosystem is N-saturated unless deposited N circumvents the terrestrial ecosystem (for example, but surface runoff). So on the face of it, it does not seem like a wise idea conceptually to try to use a standard for one ecosystem and apply it to the other. On the other hand, it could be argued that, in the absence of significant surface runoff, a linked standard is called for rather than a separate one in that aquatic ecosystems will not receive much effect of N deposition until the terrestrial system is nitrogen saturated. When it comes to S, retention in the terrestrial ecosystem will be dominated by soil chemical processes (since S limitations in terrestrial ecosystems are exceedingly rare) and negative aquatic effects will not occur until and unless 1) sulfate is mobile in sufficient amounts in soils to become a major anion in solution, and 2) soils are either naturally acidic or become acidified by atmospheric deposition. In cases where sulfate is complete mobile and soils are already acidic (for example, Spodosols in the northeast), negative aquatic effects can be expected to occur immediately (and disappear immediately as S deposition declines). These are complicated questions that will probably need to be resolved with simulations of many different scenarios and linked standards would, on the face of it, seem to be necessary.

## Dr. Naresh Kumar

### **Chapter 2: Known or Anticipated Ecological Effects**

**1. What are the views of the Panel on the appropriateness of staff's weight-of-evidence approach which assesses information from across the various ecological research areas described in the NO<sub>x</sub>/SO<sub>x</sub> Secondary Standards Integrated Science Assessment (ISA), including studies of acidification and nutrient-enrichment in aquatic and terrestrial ecosystems, and laboratory research on responses of plant and animal species to acidification and nutrient enrichment? To what extent is the presentation of evidence drawn from the ecological effects studies assessed in the ISA technically sound, appropriately focused and balanced, and clearly communicated?**

The staff has adequately summarized the information contained the ISA.

**2. To what extent are the interpretation and presentation of the results of the exposure and risk assessment technically sound, appropriately balanced, and clearly communicated?**

The staff has adequately summarized the information contained in the REA, but their communication could be improved for clarity.

Additional discussion should be provided on the assumptions and limitations of the models used in the ecological effects studies.

**3. What are the views of the Panel on the critical uncertainties associated with the risk and exposure analysis and the evidence from ecological effects studies that need to be characterized in terms of their potential implications for the secondary standards?**

The discussion on critical uncertainties (Pages 35-37) requires a more rigorous treatment than what is included in the current draft. The ranking of uncertainty (high, fairly high, intermediate or low confidence) is based on staff judgment without any explanation of how the staff arrived at those rankings.

For example, have the models and input parameters been adequately evaluated to assign high confidence in them? If so, it is essential to provide justification for the assigned rankings.

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

**10. What are the views of the Panel on staff's proposed conceptual framework for the structure of a multipollutant, ecologically relevant standard for NO<sub>x</sub> and SO<sub>x</sub>? 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 NO<sub>x</sub> and SO<sub>x</sub>?**

Figure 5-1 correctly depicts the linkages between ecological indicators, atmospheric deposition and ambient NO<sub>x</sub> and SO<sub>x</sub> concentrations. However, the uncertainties in the transformation functions between ecological response and deposition and between atmospheric deposition and air quality indicators have not been clearly explained.

For example, the Agency plans to use CMAQ model to develop relationship between SO<sub>x</sub> and NO<sub>x</sub> concentrations and the atmospheric deposition. However, CMAQ model was not adequately evaluated (either for its prediction capability for different species concentrations or for wet deposition, as a test of overall model capability) for the 2005 application that was used in the REA.

Although the data for evaluation may be sparse, it would still be useful to know how relationships between deposition and ambient concentrations obtained using the data compare against those using the model.

Given the uncertainties in the models (both atmospheric and aquatic) and the data bases, the Policy Assessment provides no indication on what ambient concentrations of SO<sub>x</sub> and NO<sub>x</sub> would meet a particular ANC. Simply stated, the Policy Assessment document does not provide information on what the confidence interval for the ambient concentrations may be (For example, is it a factor of 2, 5 or even 10).

The agency should conduct a thorough analysis of the sensitivity to different assumptions at every stage of the linkages depicted in Figure 5-1 to provide quantitative estimates of the uncertainty range in the final ambient concentration numbers needed to meet a particular level of ANC.

## **11. What are the views of the Panel on the relevance of the conceptual design for developing standards to protect against aquatic acidification effects?**

### **a) What are the views of the Panel on staff's proposed options for the ecosystem acidification model to represent the ecological response function?**

The modeling approach suggested by the staff to represent the ecological response function seems reasonable; however, the key question is how the different terms in the equation are estimated.

For example, the two approaches suggested by staff for estimating Neco give very different results and (as noted by the staff) it is not clear which approach is better.

The approach to calculate [BC] is not clearly articulated in the Policy Assessment or its appendices. Furthermore, there is a lack of any discussion about the *quantitative impact* of uncertainties in estimating [BC].

### **b) What are the views of the Panel on the relative merits of the two techniques for calculating Neco, the parameter representing the amount of deposited nitrogen that is available for acidification due to uptake, denitrification and immobilization?**

The staff should attempt to reconcile the difference in the two approaches as they give very different results. In addition, the staff should indicate how data for use in these calculations will be collected and with what frequency.

As noted earlier, both methodologies should be used as part of a sensitivity study to explore the impact of all input parameters and assumptions on final concentration values.

**c) What are the views of the Panel on staff’s proposed options for developing acid sensitivity classes to categorize the national landscape? Is it appropriate to base this classification on bedrock geology? Should multiple criteria be used to inform the sensitivity categories?**

As noted in the Policy Assessment, bedrock geology is a key factor influencing acid sensitivity, but there are additional factors. Other factors include topography, elevation, soil depth, soil composition/pH, meteorology, and land use. The assessment should explain how additional factors may influence the classification process.

**d) What are the views of the Panel on staff’s proposed options for how to aggregate acidification modeling from the catchment-scale to represent acid-sensitivity categories at the national-scale?**

The example provided in the Policy Assessment document used the 169 catchments within the Adirondacks. However, the impact of extending this methodology to the national scale is unclear. In doing so, is there not the potential for developing a standard that is more than requisite in many regions of the country?

Given that there are multiple judgments (appropriate ANC level, percentage of catchments) and multiple uncertainties in the calculations leading to the index (Neco calculation, BC calculation) and multiple uncertainties in the back-calculation to atmospheric concentrations, the Policy Assessment has a major omission in not explaining how all these factors influence the range of potential values for a secondary NAAQS (as also noted earlier).

**e) What are the views of the Panel on staff’s suggested method to account for reduced nitrogen in the deposition metric?**

The approach to account for reduced nitrogen is problematic. First, it puts the onus of improving acidification on NO<sub>x</sub> and SO<sub>x</sub> alone when reduced nitrogen (NH<sub>x</sub>, both inorganic and organic) can be quite a considerable part of acidification in many cases.

Second, the assumption that NH<sub>x</sub> deposition is constant over time is definitely wrong. Third, uncertainties associated with ammonia emissions and using CMAQ to calculate NH<sub>x</sub> deposition—as well as wet and dry deposition of SO<sub>x</sub> and NO<sub>y</sub> are high and have not been clearly explained or quantified. Although measurement data will be “blended” with model data, the overall capability of the model to represent ambient concentrations and loss terms is essential to informing judgment on the model’s overall role in back-calculating a secondary NAAQS.

## **Chapter 6: Options for Elements of a Standard to Protect Against Effects from Aquatic Acidification**

**16. What are the views of the Panel on the degree of uncertainty associated with each element of the suggested standard, e.g. the ecological indicator; the concentration to deposition ratios, reduced nitrogen, the natural background ANC, and the ambient indicator and averaging time for NO<sub>x</sub> and SO<sub>x</sub>, and its relationship to the degree of protection that could be expected from the standard? What are the views of the Panel on how to fairly characterize the uncertainty associated with the degree of protection that such a standard would provide from aquatic acidification?**

The uncertainties associated with various elements of the standard have not been clearly delineated. As a result, it is difficult to state with any degree of confidence on the degree of protection that could be expected from a given standard. As mentioned earlier, the best way to characterize uncertainty associated with the proposed approach would be to conduct a sensitivity study for each element of the standard and propagate these uncertainties to arrive at a range of protection that a given standard would provide from aquatic acidification.

**18. What are the views of the Panel on the use of regional air quality modeling (e.g., CMAQ) results to establish the concentration-to-deposition ratio (VNO<sub>y</sub>, VSO<sub>x</sub>) and reduced nitrogen deposition (NH<sub>x</sub>) in the AAPI calculation? What are the views of the Panel on the critical uncertainties associated with the use of CMAQ to generate these parameters, and the potential implications of these uncertainties for the secondary standards?**

There are a variety of problems with the use of regional air quality modeling to establish VNO<sub>y</sub>, VSO<sub>x</sub> and NH<sub>x</sub>. Although CMAQ is a regional air quality model that has been under development for many years, it lacks in its capability to correctly predict the concentrations of different species within NO<sub>y</sub>, SO<sub>x</sub>, NH<sub>x</sub> and the wet deposition of these species. 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.

A comparison of CMAQ derived values with measurements is needed before any confidence can be derived on the use of the model to generate the desired parameters.

There are many sources of uncertainties that can affect the final numbers derived from the model. These include, but are not limited to:

- large uncertainty in emissions (particularly NH<sub>x</sub> emissions, but also NO<sub>x</sub> emissions);
- inability of models to correctly predict clouds (both in cloud cover and liquid water content) and precipitation; and
- incomplete representation of chemistry (particularly cloud chemistry, nighttime chemistry and reactions with soil components).

## Dr. Myron J. Mitchell

*Responses to “Charge Questions” are provided in italics.*

### LIST OF ACRONYMS AND ABBREVIATIONS

#### Detailed Comments

<u>Page</u>	<u>Line</u>	<u>Comment</u>
x	4	LTER should be defined as “Long Term Ecological Research”—delete monitoring.
x	29-30	Delete “reduced”.
xii	30-31	The definition for dry deposition implies that dry deposition only occurs when there is not wet deposition—this is confusing. Delete “in the absence of precipitation (e.g., rain, snow) or occult deposition (e.g., fog)”.
xii	37-40	The definition of ecosystem is too broad. The earth is not an ecosystem. Delete “Ecosystems cover a hierarchy of spatial scales and can comprise the entire globe, biomes at the continental scale, or small, well-circumscribed systems such as a small pond”.
xii	7	The definition of eutrophication is not correct. Other elements (e.g., P) can result in eutrophication. Substitute “nutrients” for “nitrogen”.
xiv	4	Change to “other forms of precipitation”.

#### Introduction

#### Detailed Comments

<u>Page</u>	<u>Line</u>	<u>Comment</u>
2	6	Replace “elements” with “components”.
5	7	Why are “commercially managed forests and agricultural lands” excluded? Does this mean that those portions of the Adirondacks that are used for commercial forestry are not to be considered?
7	1	Replace “components” for elements. I would suggest the term “element” should be avoided in this document to avoid any confusion with chemical elements.

- 8            22      “m3” needs to have the 3 superscripted.
- 9            6        Change to “that SO<sub>x</sub> air concentrations have”.
- 9            19-20 This statement is not true and should be changed to “since dry deposition was believed to account for a substantial portion of the total acid deposition problem”.
- 13          4        Delete “past”.
- 15          28-29 It is not evident that emphasis has been added. Do the quotations indicate emphasis?
- 17          3-5     I agree that this review should not include those managed systems that are subject to N fertilization, but there are large areas of forest land that are managed (e.g., timber harvest) and not fertilized. These forests maybe highly susceptible to the effects of acidic deposition due to the combined deleterious effects of cation nutrient depletion due to biomass removal and leaching from the forest soil.
- 18          11      Modify this statement to be more comprehensive: “acidification, nutrient depletion and the mobilization of toxic metals in sensitive aquatic and terrestrial ecosystems”.
- 19          1        This beginning sentence shows how the term “element” or in this case “elements” can lead to confusion as indicated in my previous comments.
- 19          23      Change to “deposition decreased”.
- 19          24      Change to “deposition is decreased”. There may be confusion in terms when discussing nitrogen in the use of the term “reduced” since it also has a chemical meaning.

## **Chapter 2: Known or Anticipated Ecological Effects**

1. What are the views of the Panel on the appropriateness of staff’s weight-of-evidence approach which assesses information from across the various ecological research areas described in the NO<sub>x</sub> SO<sub>x</sub> Secondary Standards Integrated Science Assessment (ISA), including studies of acidification and nutrient-enrichment in aquatic and terrestrial ecosystems, and laboratory research on responses of plant and animal species to acidification and nutrient enrichment? To what extent is the presentation of evidence drawn from the ecological effects studies assessed in the ISA technically sound, appropriately focused and balanced, and clearly communicated?

*The current document provides a good review based upon the ISA. There needs to be clarification, however, with respect to some of the details including how the document provides results with respect to specific years. The document needs to be made more explicit with respect to the actual year(s) of the analyses. Some of the discussion is repetitive and some additional editing should be used to decrease the redundancy. Specific suggested changes are provided in my detailed comments.*

2. To what extent are the interpretation and presentation of the results of the exposure and risk assessment technically sound, appropriately balanced, and clearly communicated?

*The presentation and summarization of results related to exposure and risk assessment are generally good.*

3. What are the views of the Panel on the critical uncertainties associated with the risk and exposure analysis and the evidence from ecological effects studies that need to be characterized in terms of their potential implications for the secondary standards?

*More attention needs to be placed on the issues related to the uncertainties of the deposition estimates provided and used by various models. This is especially important with respect to estimates of dry deposition and deposition via clouds and fog. The descriptions of other issues of uncertainty are sound and include considerations of the degree of uncertainty related to different ecosystems and their respective components.*

### **Detailed Comments**

<u>Page</u>	<u>Line</u>	<u>Comment</u>
22	26	Replace “significance” with “importance”. Reserve the use of the term “significance” here and elsewhere for indicating statistical significance.
23	5	Delete “and thermodynamic processes”. This is not needed since thermodynamics is a function of the chemical reactions.
23	7	Not sure what is meant by “ecosystem exposure”.
23	13-14	Change to “and reduced chemical species”.
23	23	Replace “host” with “variety”.
23	26	Change to “but others, like geology, vary over larger spatial scales”.
24	9	Change to “episodes with deleterious consequences to sensitive biota”.
24	17	Change to “biogeochemical components”.
24	20	Change to “water bodies; moreover, deposition”. Also change “effects” to “affects”.

- 25 2 Change to “in inorganic Al concentration. These changes contribute to declines”.
- 25 4 Change to “animal species in various ecosystems. These fish may also serve as a source of food and recreation”.
- 25 15 Replace “communities” with “taxa”.
- 25 17 Change to “multiple studies are: ”.
- 25 19 Change to “20%, exchange chemistry”.
- 25 20 Change to “Under these conditions”.
- 25 23 Change to “The Ca<sup>2+</sup> and Al concentrations in soil water”.
- 26 3 Delete “episodic”.
- 26 3 Do not capitalize “sugar”.
- 26 4 Change to “cations from soil with low levels of available Ca”.
- 26 5 Change to “forests due to grassland soils being generally rich in base cations”.
- 26 9-10 This is not a very useful sentence. A more specific delineation of sensitive ecosystems is needed beyond freshwater and terrestrial. Either delete or revise.
- 26 16 Change to “flowpaths”.
- 26 16 Delete “at relatively high elevation”.
- 26 18 I would suggest that the term “magnitude” is not a very useful term. How about “range”? Also, line 21.
- 27 2 Delete “since the 1980s”.
- 27 7 I would be careful here with respect to how to phrase this description since within the western U.S. there are acidic waters associated with various factors including acid mine drainage. Maybe something like: There the acidification of surface waters by acidic deposition is uncommon in the western U.S.
- 27 22 Replace “classes” with “taxa”.

- 27 22 Change to “whereas other taxa are reduced to only acidophilic species”.
- 27 23-24 Change to “changes in taxa composition is associated with the high energy cost”.
- 28 1 Delete “In the literature,”.
- 28 7 Delete “from the EPA-administered”.
- 28 Figure 2-1 Replace “forms” with “taxa”.
- 29 1-4 In this section be more explicit of the actual date(s) related to these results.
- 29 6 Change to “there would be no improvement in water quality”.
- 29 7-8 Change to “same from 2020 to 2050”.
- 30 Figure 2-3 Figure caption. Change from “and current (2006) conditions” to “and 2006 conditions”.
- 31 5 Provide the actual year versus indicating “current”. Also (line 7) give the actual year instead of indicating “today”. Such changes should be made for the entire document.
- 32 12 Change to “emission levels from 2010 to 2050”.
- 32 15 Change to “are predicted to increase by 5%”.
- 32 17 Change to “blacknose dace”.
- 33 Figure 2-6 This figure needs a scale indicator of size. Such scales are needed in all figures showing spatial location.
- 34 11 Change to “Tree health has been linked to the availability of base cations”.
- 34 12 Change to “Tree species show a range of sensitivities to”.
- 34 13 Clarify if these ratios are “molar” or “mass” based.
- 35 15 Change to “studied tree species”.
- 35 19 Delete “in 2002”.
- 37 1 Change to “data sets”.
- 37 4 Change to “Ca<sup>2+</sup>”.

- 37 18-19 Change to “Nitrogen deposition is a major source of anthropogenic nitrogen”. For many terrestrial and freshwater systems other sources of nitrogen including fertilizer and waste treatment are greater than deposition.
- 37 18-21 The statement associated with estuaries is confusing. This needs to be reworded to indicate that for estuaries that other sources of N input greatly exceed N deposition.
- 37 29 Change to “biogeochemical”.
- 38 27 Change to “The most sensitive terrestrial taxa to N deposition”.
- 39 27 Change to “sensitive terrestrial ecosystems to N deposition”.
- 42 Figure 2-8 Delete in caption “with the inclusion of the diatom changes in the Rocky Mountain Lakes”.
- 45 22 Change to “0.7 kg N/ha/yr”.
- 46 7-8 As stated in a previous comment that the wording here with respect to the effect of N loads on estuaries is confusing.
- 46 20 Change to “eutrophication; however,”.
- 50 28 Change to “such as water availability, humidity”.

### **Chapter 3: Considerations of Adversity to Public Welfare**

4. What are the views of the panel regarding the characterizations of adversity to public welfare presented in this document? What are the views of the panel regarding the use of the ecosystem services framework as an additional metric to inform questions of adversity? What are the views of the panel regarding the usefulness of including economic valuation of some of these ecosystem services in the policy assessment document?

*The inclusion of public welfare seems to be appropriate for this document. Clearly understanding the role of ecosystem services is central to setting these standards. Hence, a description of the importance of these services needs to be a focal point of the document. The use of economic valuation also needs to be included so that the public will be aware of the issues as they relate to economic issues and other ways to evaluate how the standards affect the public welfare. Clearly a major challenge in developing the secondary standards will be to provide to the public information on ecosystem services and how maintaining these services is critical for the welfare of both individuals as well as the Nation. The more the importance that ecosystem services can be articulated, the better this document will serve in educating the public.*

5. To what extent is the presentation of ecosystem services in this document scientifically sound and clearly communicated?

*The overall description is adequate, but further editing will be needed to sharpen the focus and to be sure to avoid confusion among the various issues relating to public welfare and setting secondary standards for SO<sub>x</sub> and NO<sub>x</sub>. In some places it is not clear on the relative concerns associated with procedures to make evaluations on ecosystem services versus having the necessary information for making these evaluations. There is also an issue related to whether there should be an attempt to make an evaluation with respect to any positive aspects related to nitrogen deposition. If there is an exclusion of the determinations of the positive benefits, this might be construed as a major bias by EPA in this evaluation.*

6. What are the views of the Panel on the critical uncertainties associated with articulating adversity to public welfare that need to be characterized in terms of their potential implications for the secondary standards?

*Some further elaboration of how the types of uncertainties may affect the setting of secondary standards and how these uncertainties vary with type of effect on public welfare is needed.*

### Detailed Comments

<u>Page</u>	<u>Line</u>	<u>Comment</u>
63	2	Is this statement on limiting welfare effects of ozone to vegetation correct? Ozone can have major impacts on human health and deterioration of materials?
65	4-11	This text is very difficult to follow and needs to be rewritten.
66	4	Change to “20 mg/L” and “CaCO <sub>3</sub> ”. (See also line 7 and throughout this section where similar changes are needed) Note that the proper symbol for liters is “L”.
70	7-8	Change to something like “Especially important is the acknowledgment that it is difficult to measure and/or monetize the goods and services supplied by ecosystems”.
74		Figure 3-4 legend. Do not capitalize nitrogen and sulfur. Also the source of these data needs to be provided. Units need to be given.
75		Figure 3-5 legend. Do not capitalize nitrogen and sulfur. Also the source of these data needs to be provided. Units need to be given.
75-76		Change the ending and beginning sentence of each of these pages to something like: “The difficulty in the monetization for ecosystems services has

been previously emphasized hence necessitating using a subset of services in economic valuation.

- 76 14 Delete “colloquially”.
- 76 14 Change to “However, the amount an individual is willing to pay”.
- 77 21 Do not capitalize “bald eagle”.
- 78 2 Including the term “costly” confuses issues related to evaluation of public welfare and monetary evaluation of ecosystem services. A major challenge in this document and for moving secondary standards forward will be to provide to the public information on the importance of ecosystem services and how the secondary standards help to protect these services.
- 79 4 Replace “certain” with “specific”.
- 79 10-11 Is it really true that food is generally the most important provisioning service provided by inland service waters? Isn’t the availability of potable water by far the most important service?
- 80 9-10 This statement is confusing with respect to whether surface waters are affecting versus being affected by hydrological regimes and climates.
- 80 28 Change to “resulting from the decrease of anthropogenic”. Isn’t it impossible to actually eliminate the anthropogenic emissions of NO<sub>x</sub> and SO<sub>x</sub>?
- 81 2 See previous statement with respect to issue of emissions being eliminated.
- 81 14-16 Change to: These model runs assumed a 2010 implementation of “zero-out” emissions with a projected lag time to improvement of 10 years and thus results were calculated for the year 2020.
- 82 6-7 Change to “indicate that impaired lakes would decrease from 22 to 31% using background conditions”.
- 82 11-15 This sentence is very confusing and needs to be reworded.
- 82 16-17 Change to “This analysis provides results on only a subset of the impacts of acidification on public services and suggests that the overall impact on these services is likely to be substantial”.
- 82 22 Clarify whether the issue is that there is no known procedure to make these determinations or whether we do not have the necessary information.

- 82 29 Do not start new paragraph.
- 82 30 Change to “producing timber and maple syrup that”.
- 83 6 Give the names of these two listed species.
- 83 16 Replace “roughly” with “approximately” here and throughout document.
- 84 3 Replace “regulate that quantity and flows” with “help regulate the quantities and temporal discharge patterns”.
- 84 3-4 Delete the sentence starting with “Finally” and replace with “Forests also play an important role in carbon sequestration at both regional and global scales”.
- 84 24-29 This paragraph might be interpreted that EPA is not making a full evaluation and is biasing the analyses with consideration of only deleterious effects. Better justification is needed on why the beneficial effects are not to be considered in the evaluation. Is there a problem with availability of data or is there some other important issue that prevents this analysis?
- 85 2-3 Change to “Estuaries in the eastern United States are important for fish and shellfish food production”.
- 85 20 Change to “Mistiaen et al. (2003)”.
- 86 13 Change to “\$2 billion”.
- 86 13 Change to “Almost seven million people”.
- 86 17 Indent starting new paragraph.
- 87 1 Change “air” to “atmospheric”.
- 87 3-10 Clarify if these amounts are based upon an apportionment of the relative role of atmospheric deposition to total nitrogen inputs to the Chesapeake Bay.
- 88 21-22 Provide the names of these species.
- 88 30 Correct to “deposition”.
- 89 19-20 As indicated previously, clarify if the issue is the lack of methods to make the assessment or the absence of data needed for making evaluations of the impacts on services.
- 100 Here and elsewhere in the document there is specific reference to “staff”. This suggests that there is a subcomponent of EPA that supports the narrative

within this document. I would avoid the use of this designation to avoid any confusion.

100            13      Change to “Chapter”.

#### **Chapter 4: Addressing the Adequacy of the Current Standards**

7. What are the views of the Panel on staff’s assessment of the adequacy of the form of the existing NO<sub>x</sub> and SO<sub>x</sub> secondary standards? To what extent does the Panel agree with staff’s assessment of the protection provided by existing standards, given the current levels, forms, averaging times, and indicators?

*The document provides a good review of existing secondary standards and shows that they are not adequate for ecosystem protection.*

8. What are the views of the Panel on the time frame of ecological response related to current deposition? The adequacy evaluation relies on recent NO<sub>x</sub>, SO<sub>x</sub>, deposition, and on long-term steady state ANC. Does the panel agree that long-term steady state ANC is the most appropriate representation of ANC for evaluating the adequacy of the current standards?

*The use of long-term ANC has merits with respect to setting standards, but may not adequately capture some of the temporal changes that will likely occur in conjunction with changes in surface water chemistry. Clarification is needed on the specific time to be used as targets for recovery and the actual rates of these recoveries.*

9. To what extent are the characterizations of ambient air quality and deposition appropriately characterized, relevant to the review of the secondary NO<sub>x</sub> and SO<sub>x</sub> NAAQS, and clearly communicated?

*A more balanced approach is needed in which it is more clearly delineated what information the current networks currently provide and what information is lacking. For example the network distribution may be completely adequate for estimating wet only sulfate deposition, but the estimates for NH<sub>x</sub>, including both wet and dry, are lacking. Maybe the inclusion of a figure or table that provides a description of the both spatial and temporal coverage of the major chemical species atmospheric concentrations would be useful. Some of the discussion implies that the CMAQ model will be used to overcome some of the problems with respect to spatial coverage of atmospheric pollutants and deposition of sulfur and nitrogen compounds. Within the document the limitations of CMAQ need to be provided with respect to how well the model outputs have been validated both with respect to temporal and spatial patterns.*

#### **Detailed Comments**

Page            Line    Comment

- 101 15-17 Change to “Acidification occurs over extended periods and the ability of both terrestrial and aquatic ecosystems to recover is dependent upon not only the decrease in acidic deposition, but the ability of these ecosystems to generate cations that are needed for nutrients and base cation supply”.
- 101 24 Change to “This type of structure does not take into account the spatial and temporal variability of deposition and ecosystem processes with respect to the effects of NO<sub>x</sub> and SO<sub>x</sub> on public welfare”.
- 102 2 Change to “including geological and soil characteristics related to the sensitivity to acidification as well as atmospheric and landscape characteristics that govern rates of deposition”.
- 104 10 Change to “of secondary standards”.
- 104 11 Change to “impact both nitrogen and sulfur acting”.
- 104 22 Change to “relevant chemical species”.
- 104 24 Change to “total nitrogen and total sulfur deposition”.
- 105 26-27 This general phrase has been used elsewhere in the document and does not adequately describe the issues. There are three major factors that all need to be considered: 1) Atmospheric concentrations 2) Deposition velocities of various chemical species and 3) the processing within ecosystems. It is not clear what are “atmospheric factors” and what are “ecological factors”. Is the type of vegetation (that affects deposition velocities) an atmospheric or ecological factor? Is surface wetness an atmospheric factor”? Are hydrological pathways ecological factors?
- 106 23-25 This statement is too pessimistic and suggests that the monitoring networks are completely inadequate. It would be better to state that certain components (including NH<sub>x</sub>) are not adequately measured within the current networks. Figures 4-1, 4-2 and 4-3 suggest that the network coverage is quite extensive.
- 107 2-3 In Figure 4-1 legend give the actual chemical N species for which concentrations are monitored.
- 108-111 This section provides a narrative on what chemical species the various networks monitor, but it is not easy to use this information to evaluate the adequacy of the monitoring. Some summary figures or possibly a table showing what actual chemical species are monitored and the number of sites doing this monitoring would be more helpful in evaluating the adequacy of at least the spatial coverage of the networks for specific chemical species.

- 108 2-3 In Figure 4-2 legend give the actual chemical S species for which concentrations are monitored.
- 111 16-17 This statement suggests the CMAQ model will satisfy the needs with respect to providing data on atmospheric concentration and deposition of S and N chemical species. The limitations of the CMAQ need to be acknowledged.
- 111 29 Do not capitalize “organic”.
- 111-114 The discussion on the rationale with respect to the use of CMAQ is useful, but little quantitative results are provided with respect to clearly indicating what CMAQ does well and where are the problems.
- 114 18-22 The focus of CMAQ on relatively short time (e.g., hourly values) may result is a mismatch to the needs for a secondary standard that would more likely result in values needed for longer periods (e.g., yearly values).
- 115-123 These various figures show the modeled results from CMAQ for various atmospheric chemical species and show the spatial detail available from the CMAQ output. There is discussion on how these modeled values show the spatial distribution of these chemical species for the single year of simulation (2005). However, this is not very helpful in ascertaining the accuracy and precision of the CMAQ simulations.
- 116 6-7 The assertion that the modeled and observed values are similar needs to be backed up with quantitative evaluations.
- 116 16-17 In discussing these conditions provide the actual year of comparison. Also, the wording may be confusing with respect to the current secondary standards and the approach being developed within the current document that is trying to develop secondary standards showing broader ecological effects.
- 125 Showing the results from CASTNET in Figures 4-12 and 4-13 is helpful, but it would be more instructive is there were direct comparisons between the CMAQ and CASTNET results.
- 126-127 For Figure legends, correct the capitalization of units.
- 132 4-5 I don’t believe that there is sufficient evidence between S deposition and Hg methylation. There is known relationship between sulfate reduction and Hg methylation, but there is no direct linkage with S deposition.
- 132 23 It would be helpful to add a third category: nutrient depletion. This category is related to acidification, but focuses on the importance of the loss of nutrient cations from ecosystems.

- 134 5 The MAGIC model certainly provides information, but there other sources of information that also indicate the importance of aquatic acidification in the Adirondacks and Shenandoah National Park.
- 134 24 Change to “concern); however”.
- 135 4 change to “ results based upon the EMAP”.
- 135 9 Here and elsewhere instead of using the term “recent”, give the actual year or period of coverage.
- 135 12-13 Change to “As for the Adirondacks, these results suggest that a substantial proportion of streams”.
- 135 28 Change “kilometers” to “lengths”.
- 136 2 Change to “two to three times greater than the number of lakes classified as”.
- 137 2 Change to “The REA did not evaluate all possible sensitive regions, but focused on specific case studies”.
- 137 2 Change to “For example, in the sugar maple case”.
- 137 14 Change to “For red spruce, 5% of all plots”.
- 137 15-16 Change to “In those states where red spruce is an important tree species (Maine”.
- 138 10 The statement “In certain limited situations” is not true. For many if not most terrestrial ecosystems N additions results in increased plant growth. This includes both natural and managed ecosystems.
- 138 14 I have commented previously on the importance of considering both “managed” and “unmanaged” systems especially with respect to forests.
- 138 20 Note that this statement is correct and conflicts with line 10.
- 138 23 Be careful in providing these numbers for nitrogen uptake by crops. Are these actual uptake values for N or the amount of N applied as fertilizer?
- 138 26 This statement is not true. There is a substantial body of information on the effects of atmospheric deposition of N to forests and other ecosystems.
- 139 21 Change to “capacity of each county’s”.

140	12	Change “were” to “was”.
140	26-31	This paragraph suggests that the standard needs to focus on these systems which are most sensitive to increases in N deposition.
141	4	Delete “scientifically”.
141	5	Change “country” to “U.S.”.
141	13	Change to “received”.
141	21	Delete “, the authors estimated that”.
141	28	Shouldn’t this be “systems where N is limiting”?
142	13-16	Is this really a link with S deposition or is this a function of sulfate concentration which can be affected by other factors in addition to S deposition?
142	29	Change to “Assessment document”.

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

10. What are the views of the Panel on staff’s proposed conceptual framework for the structure of a multipollutant, ecologically relevant standard for NO<sub>x</sub> and SO<sub>x</sub>? 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 NO<sub>x</sub> and SO<sub>x</sub>?

*Some of the discussion with respect to ecological responses and various watershed attributes were confusing in the different treatments of nitrogen and sulfur. A treatment that shows more clearly the biogeochemical similarities and differences would strengthen the overall approach.*

11. What are the views of the Panel on the relevance of the conceptual design for developing standards to protect against aquatic acidification effects?

*The overall conceptual design is adequate.*

a) What are the views of the Panel on staff’s proposed options for the ecosystem acidification model to represent the ecological response function?

*There are some concerns related to the application of this model with respect to accurately capturing the critical biogeochemical processes. There will be important tradeoffs between keeping the model relatively simple and capturing important spatial and temporal patterns affecting acidification.*

b) What are the views of the Panel on the relative merits of the two techniques for calculating  $N_{eco}$ , the parameter representing the amount of deposited nitrogen that is available for acidification due to uptake, denitrification and immobilization?

*The use of  $N_{eco}$  formulation that needs estimations of N retention, immobilization and denitrification may be very problematic since the parameter estimates (especially immobilization and denitrification) are very difficult to obtain. It would be better to focus on the actual N losses via leaching. This latter value is easier to obtain, but may require a more empirical approach. An issue that needs consideration is the time units used in these calculations. It will probably be important to include some seasonal effects since there are marked changes in nitrate losses during different periods of the year.*

c) What are the views of the Panel on staff's proposed options for developing acid sensitivity classes to categorize the national landscape? Is it appropriate to base this classification on bedrock geology? Should multiple criteria be used to inform the sensitivity categories?

*It is not appropriate to only use bedrock geology for assessing the potential for acidification and/or recovery from acidification. The surficial geology including the parent material and hydrological flow paths can be extremely important and catchments with the same parent material can show very different responses to acidification. Yes, multiple criteria are needed to provide sufficient information for sensitivity categories. There has been discussion related to the use of elevation as a predicting variable. Clearly elevation is a proxy for the more direct controls such as soil depth, vegetation type, etc. and it would be preferable to actually include the controlling variable in developing the model. At a minimum the issue of elevation being a proxy for controlling physical, chemical and biological components needs to be provided.*

d) What are the views of the Panel on staff's proposed options for how to aggregate acidification modeling from the catchment-scale to represent acid-sensitivity categories at the national-scale?

*The aggregation needs to be done in a meaningful manner that takes into account the controlling factors of acidification of surface waters. There is an over dependence on the use of bedrock geology that does not consider the importance of other critical factors, especially superficial geology including soil depth and soil chemistry, in affecting sensitivity to acidification.*

e) What are the views of the Panel on staff's suggested method to account for reduced nitrogen in the deposition metric?

*The assumption that the proportion of reduced N is constant over time, may cause a number of problems both for forecasting and also for any attempt to examine model performance using hindcasting.*

12. What are the views of the Panel on the relevance of the conceptual design for developing standards to protect against terrestrial acidification effects? Terrestrial nutrient enrichment effects? Aquatic nutrient enrichment effects? Does the panel have suggestions on additional data or methods that might enable EPA to expand the current aquatic acidification approach to cover additional effects?

*Developing a single standard for either aquatic or terrestrial ecosystems is highly problematic due to the major differences in acidification and nutrient response among and within the regions. The standard needs to vary to account for differences in the biogeochemical processes among different responses. For terrestrial systems it will be particularly important to look at different biological species including trees that have very different requirements for base status and nutrients (e.g., red spruce versus sugar maple). There are importance differences in the landscape features across the nation and the historical inputs of S and N that influence ecosystem responses including acidification and nutrient effects.*

13. What are the views of the Panel on the critical uncertainties associated with the conceptual design of an ecologically relevant multi-pollutant standard that need to be characterized in terms of their potential implications for the secondary standards?

*The development of some careful analyses related to the precision and accuracy of the CMAQ simulations needs to be included. It would be helpful to also show any other regional approaches that have estimated N and S deposition and how these compare with CMAQ simulations.*

14. To what extent do the figures and examples aid in clarifying the text? Should more or less information of this type be included in the second draft?

*The figures and examples need to be modified to capture the major processes that control ecosystem response to acidification and nutrients. See specific comments for more details.*

### **Detailed Comments**

<u>Page</u>	<u>Line</u>	<u>Comment</u>
147	9	Change “big” to “major”.
148	9	The term “landscape features” is rather vague. Does this include physical, chemical and biotic attributes? Isn’t there an important issue related to the history of deposition for given areas with the most sensitive areas to N deposition in the west which have been subject to relatively low atmospheric N inputs?
148	11	How is the country being subdivided? Are these states, biomes, ecosystem types, or something else?
149	19	Delete “scientific evidence”.
149	20-22	Change to “aquatic acidification should be measurable and causally linked to the deposition of N and S. The indicator should have ecological effects that adversely affect public welfare”.

- 150        6        Change to “impairment, including the number of fish species”.
- 150        15        Change “acid anions” to “mobile anions”.
- 150        16        Change to “when these anions are mobilized in”.
- 150        22        Change to “deposition. These acidification models simulate a variety”.
- 150        26        Change to “ANC= 50 :eq/L”.
- 151        22        Change to “These data were”. Change “current” to “recent”.
- 152        3        Delete “time”.
- 152        15        Change to “with different levels”.
- 152        24        Change to “due to variation in biogeochemical processes among watersheds”.
- 152        25        Change to “that quantifiable relationships exist”.
- 152        26-27    Change to “These relationships are shown by long-term”
- 152        28-29    Change to “Models are important tools in evaluating how”.
- 153        3        Change to “There are various factors that modify”.
- 153        4        Change to “of these factors are described”. Delete “that parameterize ecosystems to simulate the process”.
- 153        6        Change to “input data for 17 to 20 environmental”.
- 153        9        Change to “specific responses of aquatic ecosystems”.
- 153        12-13    Change to “The weathering of soil minerals is a major source of base cations to ecosystems”. (Note this it may not be the main source in all cases.)
- 153        15        Change to “Landscape factors that affect the acid sensitive of forest ecosystems”.
- 153        19-20    Delete from “Numerous ..... example,”.
- 153        22        Change to “Water discharge values for the catchment”.
- 153        27        Replace “quality” with “chemistry”.

- 154 8 Replace “correlated” with “linked”.
- 154 8-9 Delete the sentence “Consideration....models”.
- 154 10-11 Change to “The calibration of the models stream water chemistry, soil characteristics and atmospheric deposition estimates”.
- 154 15 Change to “will vary among catchments”.
- 154 16-17 Delete the sentence “However.... simulations”.
- 154 20 Replace “quality” with “chemistry”.
- 154 24 Give the units for the ANC values.
- 155 8 Delete “mountain”.
- 155 14-16 Change to: “With these data the atmospheric deposition loads that will cause streams to exceed the critical limit of ANC can be calculated”.
- 155 18 Delete the sentence “The relationship....nationally”.
- 155 19 Change to “similar hydrology and mineral weathering rates should show similar”. Need to be careful in these statements since the amount of base minerals generated is a function not only the weathering rates of soil minerals, but also the hydrological relationships including flow paths.
- 155 21 Is there an implicit assumption of what maximum area constitutes a “catchment”?
- 155 24-26 This statement is confusing in that critical load is highly dependent on the characteristics of a catchment. How can aggregation be used to develop a national standard? Isn’t it more likely that different standards will be needed for different catchments or that standards will be set to protect a certain subset of catchments?
- 156 3-7 It is not clear if developing a single standard is the goal of this process. See my previous comment.
- 156 14-15 Change to “Models are important tool for evaluating how multiple”.
- 156 17 Change to “Mineral weathering is a major source of base cations and is therefore considered an important factor in determining critical loads”.
- 156 19 Replace “correlated” with “linked”. Here and elsewhere the use of the term correlated is somewhat weak since this infers there is a statistical correlation

and not necessarily a causal relationship. Using factors that have casual relationships is a stronger approach than using statistical correlations.

- 156 21 Delete sentence “Modeling every...requirements”.
- 156-157 These summary statements should clarify that having a single secondary standard may not be warranted to take into account the substantial variation in the sensitivity among regions with respect to surface water acidification.
- 157 4 Replace “to” with “for”.
- 157 7 Change to “to protect different areas”.
- 157 13 Delete the phrase “scientific literature”. Here and elsewhere in the document it is implicit that the overall procedure will be scientific and this phrase is not needed.
- 157 20-23 This dichotomy is not valid since S can certainly be retained in ecosystems especially those with high potential for sulfate adsorption. Both the leaching of nitrate and sulfate as well as the retention of N and S need to be part of the overall evaluation.
- 157 26 This is confusing since there is generally little ammonium lost in runoff in systems sensitive to acidification. Maybe this should be restated with respect to N solute leaching and hence the inclusion of DON would also be warranted.
- 157 16 The legend for Figure 5-3 needs more information including the meaning of the various abbreviations.
- 159 1-23 The use of these models and the different treatment of S and N is confusing. It would be clearer if the models more accurately captured the biogeochemistry of each of these elements. One issue that could be important in some of the areas is that there is a net loss of sulfate from soil that is greater than atmospheric inputs and this will delay the recovery of these systems from acidification.
- 160 8-10 The use of  $N_{eco}$  formulation that needs estimations of N retention, immobilization and denitrification may be very problematic since the parameter estimates (especially immobilization and denitrification) are very difficult to obtain. It would be better to focus on the actual N losses via leaching. This value is easier to obtain. An issue that needs consideration is the time units used in these calculations. It will probably be important to include some seasonal effects since there are marked changes in nitrate losses during different periods of the year.

- 160 12 As indicated previously the assumption that all S deposition is leached is not valid.
- 161 9 The goal of “obtaining a representative deposition value” is very problematic due to the high variation in acid base chemistry response among catchments is very high (e.g., even adjacent catchments may have very different responses).
- 161 12-23 There may be merit in the identification of specific population of surface waters that have substantial representation among all water bodies of a region, but are most susceptible to acidification.
- 162 3-10 The use of bedrock geology for the classification of acid sensitive catchments is problematic. The surficial geology needs to be used in this classification. In the early days of “acid rain” research the use of bedrock geology was used in classifying watersheds, but as the understanding of acidification processes increases it became apparent that surficial geology including soil depth is very important in evaluating spatial variability of sensitivity to acidification. For example using Figure 5-4 it is indicated that the central Adirondacks would be most sensitive to acidification, but we know that the highest proportion of acidic surface waters in the Adirondacks are found in the southwest portion of this region.
- 163 6-15 Here and elsewhere in the document the use of elevation as a parameter is suggested in the analyses. Elevation is not the casual factor associated with sensitivity to acidification, but it is related to factors that are correlated with elevation including precipitation amount, soil depth, changes in vegetation, etc. It would be better to use the actual causal factor in these determinations or at least indicate that elevation is linked to these causal factors.
- 164 25-27 The statement that “reduced forms of nitrogen deposition” are quickly converted to nitrate is not correct. Some reduced forms of N can be assimilated by both the vegetation and the soil microflora without being converted to nitrate. If nitrification rates are low N deposited in reduced forms may also not be converted to nitrate. It should be emphasized that the total amount of N deposited is the factor that determines the N atmospheric loading. This is why total N deposition needs to be considered in making these evaluations.
- 165 2-3 Isn’t the most important assumptions associated with any temporal changes in N deposition versus the relative importance of oxidized versus reduced forms of N deposition.
- 165 7-29 Within this “summary” a number of statements related to the use of bedrock geology, deposition calculations, etc. for which concerns have been provided in comments above.

- 166 Table 5-2 (this is actually a figure versus a table). The descriptors should be changed to more clearly reflect the factors that influence acidification.
- 167 21 “This leads to the question, are aquatic or terrestrial ecosystem more sensitive?”—aquatic and terrestrial ecosystem responses to acidification differ with the effect of limiting nutrients (e.g., calcium) more important for terrestrial versus aquatic ecosystems. This actually is implicit in the use of different indicators (e.g., Bc:Al for terrestrial and ANC for aquatic systems, respectively).
- 168 11 It is really feasible “one indicator that can be applied across the nation”. This may not be possible with different conditions among regions (e.g., west having lower historical rates of atmospheric N inputs).
- 169 2 Change to “first draft PA; however, a”
- 169 1-19 Concerns related to these summary statements are provided in my previous comments.
- 169 22-24 Clarification is needed on how cloud/fog deposition fits into these two mechanisms.
- 169 25 Change to “and chemical species-specific”.
- 169 10 See above comment associated with incorporation of fog/cloud deposition into this formulation.
- 171 1-2 The rationale for using  $V_{S/N}$  is not evident due the great variation in deposition velocities among various atmospheric chemical species.
- 171 2 Change to “aggregated deposition”.
- 171 4 Change to “the chemical species specific”.
- 171 8 Clarify what is meant by “annual average”—isn’t the relative value the total annual deposition?
- 171 13-15 It is not clear what is being suggested by this statement.
- 171 25-27 The development of uncertainties in the CMAQ predictions will be extremely important in understanding the accuracy and precision of these estimates.
- 171-172 The procedure for calculating  $V_{S/N}$  seems very convoluted and it is not evident how this is better than calculating separately annual deposition of N and S.
- 178 3-4 Correct units.

- 179 2-3 As stated previously, the calculation of a depositional load for a specific ANC that does not account for variation in catchment responses to acidification will not result in accurate predictions of how these systems are responding. At least some classification of catchment types is needed. Such classifications have been used extensively in some of the regions such as the Adirondacks (e.g., (1) mounded seepage lakes with low levels of DOC; (2) mounded seepage lakes with high DOC; (3) drainage lakes in watersheds with thin till and with low DOC; and (4) drainage lakes in thin-tilled watersheds with high DOC.
- 180-188 Before developing further details on these calculations, agreement is needed with respect to the overall approach. Many of these calculations are based on the need to develop a single standard. Are there other approaches that should be considered?

## **Chapter 6: Options for Elements of a Standard to Protect Against Effects from Aquatic Acidification**

15. To what extent does the Panel agree that the proposal to develop an Atmospheric Acidification Potential Index (AAPI) linked to key determinants of aquatic ANC is a reasonable approach to developing an ecologically relevant standard? Does the Panel generally agree that the secondary standard options identified by staff (including indicator, averaging time, form, and level) are generally consistent with the available scientific and technical information and are appropriate for consideration by the Administrator?

*The Atmospheric Acidification Potential Index (AAPI) is an interesting approach that integrates aquatic ANC as affected by acidic deposition. The use of this index needs to be considered with respect to issues associated with temporal and spatial variability. Although the current approach allows for the development of unified national standards, it does not account for the high level of biogeochemical variation in the processing of nitrogen and sulfur in catchments. Especially for those catchments which have major differences in seasonal drainage rates (including snowmelt and droughts) time scales are needed that will reflect episodic conditions. There is a large body of evidence of the importance of the spatial and temporal variation in reflecting acidification and nutrient enrichment processes among ecosystems. Some specific examples showing how these standards would account for episodic responses would be very helpful in showing that the standard would protect those systems for which episodic acidification is the major issue. This would help link this to climate change effects on hydrology and biogeochemistry. The sensitivity to episodic components could also be related to watershed type distributions including the presence of wetlands that would be especially important for episodes associated with rewetting after droughts.*

16. What are the views of the Panel on the degree of uncertainty associated with each element of the suggested standard, e.g. the ecological indicator; the concentration to deposition ratios, reduced nitrogen, the natural background ANC, and the ambient indicator and averaging time for

NO<sub>x</sub> and SO<sub>x</sub>, and its relationship to the degree of protection that could be expected from the standard? What are the views of the Panel on how to fairly characterize the uncertainty associated with the degree of protection that such a standard would provide from aquatic acidification?

*More attention is needed on other factors that will affect the biogeochemical processing of N and S within watersheds. Also, the strong reliance in using the CMAQ model would be greatly improved by more specific comparisons related to actual comparisons between CMAQ estimates and measured values. Consistency is needed between the temporal and spatial resolution of the depositional predictions and the actual measurements within terrestrial and aquatic ecosystems regarding nutrient enrichment and acidification.*

17. What are the views of the Panel on aggregating the terms (Q, N<sub>eco</sub> and BC0 \* ) used in estimating natural background ANC, denoted by the function g(A), into 5 bins based on the geologic classification scheme?

*I don't believe the use of the geologic classification scheme is adequate to capture the spatial and temporal patterns needed for setting these standards. This can be shown by looking at the modeled outputs in the Adirondacks that do not appear to show what is known about the spatial patterns of surface water acidification in this region. At a minimum more attention is needed with respect to surficial geology and soil properties both with respect to physical aspects (e.g., soil depth) and chemistry (e.g., base cation concentrations).*

18. What are the views of the Panel on the use of regional air quality modeling (e.g., CMAQ) results to establish the concentration-to-deposition ratio (VNO<sub>y</sub>, VSO<sub>x</sub>) and reduced nitrogen deposition (NH<sub>x</sub>) in the AAPI calculation? What are the views of the Panel on the critical uncertainties associated with the use of CMAQ to generate these parameters, and the potential implications of these uncertainties for the secondary standards?

*As indicated in previous comments there are a number of issues related to the use of the CMAQ results. A critical component of this analysis is the determination of the precision and accuracy associated with the calculation/estimates of deposition velocities of the chemical species of sulfur and nitrogen.*

19. What are the views of the Panel regarding presentation of the standards as a set of tradeoff curves for NO<sub>x</sub> and SO<sub>x</sub> associated with specific levels of AAPI, and specific values of the g, VNO<sub>x</sub>, VSO<sub>x</sub>, and NH<sub>x</sub> terms?

*To make valid assessments of the effects of N and S deposition on ecosystem processes the various components that provide the deposition and the actual chemical species (especially for nitrogen compounds) need to be delineated separately.*

20. What are the views of the Panel on using a single year or a three year average of recent year CMAQ modeling results to estimate the AAPI terms?

*The use of such a short time frame for developing terms should be reconsidered especially in the context of ecosystem level responses that occur over much longer periods (e.g., decades and longer), but having multiple years for comparison would be helpful in showing how meteorological conditions affect or do not affect the AAPI calculations. Is there an issue with respect to the limitations of results from the CMAQ model in determining what information is available with respect to atmospheric deposition?*

21. What are the views of the panel on the ambient monitoring requirements? Is for the proposed three ambient air measurements – NO<sub>y</sub>, SO<sub>2</sub> and particulate sulfate – sufficient to judge compliance with and AAPI?

*It is not completely clear whether this approach (especially aggregating information on watershed responses) will result in the development of useful results for predicting the spatial and temporal patterns of acidification and most importantly how these systems will respond to changes in loads of atmospheric deposition. This is also important with respect to the watershed response monitoring components both biological and chemical.*

22. What are the views of the Panel on using existing NO<sub>y</sub> and PM<sub>2.5</sub> sulfate measurement techniques as the basis for defining a Federal Reference Method to judge compliance with and standard?

*There are important issues with respect to whether NO<sub>y</sub> captures the major N chemical species. For PM<sub>2.5</sub> the amount of S is relatively high in this atmospheric fraction, but its overall contribution to S loading is relatively small; hence this is not the major issue. For S the major problem is associated with making accurate and precise estimates of S in dry deposition and also for some sites deposition from fog and/or clouds.*

### **Detailed Comments**

<u>Page</u>	<u>Line</u>	<u>Comment</u>
190	15-16	I agree that the previous methods for defining indicator, averaging time, form and level are not appropriate with respect to ecosystem level effects associated with N and S deposition.
190		The statement that: “Moreover, the inherently complex and variable linkages between ambient concentrations of NO <sub>x</sub> and SO <sub>x</sub> , their deposited forms of nitrogen and sulfur, and the ecological responses that are associated with public welfare effects call for consideration of a more complex and ecologically relevant design of the standard that reflects these linkages” needs to be reflected in the procedures used to develop these standards.
191-192		Section 6.1 seems to be highly redundant with respect to information provided in previous chapters.

- 193-194 Section 6.2--I agree that future efforts will need to consider how to incorporate results associated with episodic versus chronic responses. This will be especially important in those areas in which snowmelt is a dominant component of the hydrologic cycle.
- 194-195 The development an Atmospheric Acidification Potential Index (AAPI) has some potential, but this formulation must be sufficiently robust to account for differences in biogeochemical responses among watersheds.
- 195 15 I agree of the importance of including “including landscape and atmospheric factors” in developing these formulations.
- 196 As stated previously, much of this discussion and formulation is based upon the development of an aggregated value. This approach may result in erroneous recommendations if spatial variability in watershed components and historical differences in deposition are not included.
- 198 6 The other ecosystem variables will be very important with respect to predicting sensitivity to acidification.
- 200-201 The selection of the populations of areas to be considered with respect to sensitivity parameters will be important in developing these predictions. Clearly defining which subset of the overall population is sensitive to acidification and the relative importance of these sensitive populations should be a focal point of the analyses.
- 201 15-16 The need to exclude “water bodies that are naturally acidic” is a clear example of the need to account for the differences within and among regions with respect to biogeochemical responses of acidification.
- 201 20-27 The next draft should explore in addition to alternate combinations of the target ANC, but also the known variation in watershed processing of N and S.
- 203-207 Section 6.5 provides further elaboration (See also Chapter 5) of the rationale for using ANC as an ecological indicator for the acidification of aquatic ecosystems.
- 207 12-13 The time issue with respect to recovery from acidification needs further elaboration and how the identified approach will address this issue. The time issue needs to be included in earlier discussion related to the application of critical loads.
- 208 Section 6.6 covers some of the issues related to the more complete evaluation of the various atmospheric chemical species of N and S compounds. With the exception for the need for better quantification of ammonia, the most pressing issues relate to the estimates of dry deposition especially the utilization of deposition velocities.

- 208-209 Section 6.6.2 considers the issues around sampling frequency. For looking at ecosystem level effects it will be important to have extended periods of measurement. In addition to monitoring current and future concentrations, there is a strong need to have historical measurements of concentration. Having this historical information is critical for looking at long term watershed responses including modeling efforts.
- 209 27-28 Clarify what is meant by “largely population oriented”.
- 210 1 Not sure of the point indicated by “Ambient monitoring at every watershed may be required due to the nature of the ambient air quality in acid sensitive areas”.
- 210 4 I would not agree that using annual averages dampens “much of the spatial variability”. Do you mean variability of deposition estimates only?
- 210 6 How will CMAQ concentration values “provide insight into the likely spatial representativeness of monitors” without clear documentation that CMAQ is providing accurate and precise values of atmospheric concentration within region at various spatial scales.
- 210 8 How was this spatial variation of oxidized nitrogen in the Adirondacks derived? The value of 1.46% for a coefficient of variation seems very small in reflecting changes in oxidized nitrate values across this region.
- 210 Section 6.7 focuses mostly on the justification of the use of ANC and AAPI. It is not clear whether other parameters need to be considered such as the effect of N addition in sensitive aquatic and terrestrial ecosystems.

## Chapter 7: Co-protection

23. What are the views of the panel on the approach taken to compare the protection provided by a potential aquatic acidification standard to the protection needed for terrestrial acidification?

*The results presented that show differences in protection between aquatic and terrestrial systems are interesting and shows both the linkages, but also some of the problems. There are issues beyond acidification with respect to terrestrial ecosystems recognizing that soil acidification is a natural process especially in forested ecosystems and hence the major concern is related to nutrient imbalances some of which is captured by the use of the Bc: Al ratio, but for this to be meaningful this ratio needs to address the spatial variability of soil chemistry.*

24. What are the views of the panel on a future comparison of the protection provided by a potential aquatic acidification standard to nutrient enrichment benchmarks? What are the views

of the panel regarding using a nutrient enrichment benchmark to be a limiting factor on the nitrogen in the aquatic acidification standard, instead of having a separate standard?

*The section on nutrient enrichment benchmarks is not yet developed fully in the document and hence comments would be premature.*

### **Detailed Comments**

<u>Page</u>	<u>Line</u>	<u>Comment</u>
213		Section 7.1--In further development of this comparison between terrestrial and aquatic systems a clarification of the relative importance of base cation generation and hence improvement of the Bc:Al ratio in terrestrial ecosystems is linked to ANC recovery in surface waters.
214		Section 7.2--As the role of nutrient enrichment becomes more developed in this analysis this could have a marked effect on the overall approach in developing these standards especially with respect to difference among regions associated with the history of N deposition.
215		Section 7.3--The aquatic nutrient enrichment is not only associated with N effects on estuary eutrophication, but also on changes in the biota in surface waters in the western U.S.

### **Chapter 9: Conclusions**

25. What are the views of the Panel on the preliminary staff conclusions regarding the adequacy of the current standards, the need for an integrated multi-pollutant structure for revised standards, and the proposed form of the joint NO<sub>x</sub> SO<sub>x</sub> standards for aquatic acidification?

*The review of current standards and the need for an integrated multi-pollutant structure for revised standards is well done and summarizes the importance of this effort for developing new secondary standards.*

26. What are the views of the Panel on the overall characterization of uncertainty as it relates to the determination of an ecologically-relevant multi-pollutant standard for NO<sub>x</sub> and SO<sub>x</sub>?

*The section on uncertainty is yet to be fully developed and this will likely be separate chapter. The determination and the discussion of uncertainty in all aspects of the development of the secondary standards including atmospheric inputs and ecosystem effects should be a predominate component of this document.*

### **Detailed Comments**

<u>Page</u>	<u>Line</u>	<u>Comment</u>
218	19-20	The current focus on acidification is well justified.
219-222		The conclusions indicate the major findings of this review especially the rationale for focusing on acidification in the current version.
222	20	A brief summary that defines AAPI should be provided in the summary.
223		The section yet to be written on key uncertainties will be an important component of this report and associated recommendation. It will likely be necessary to include the effects associated with nutrient alterations that is a critical issue for both the east (Ca depletion in terrestrial ecosystems) and west (N deposition affects on ecosystem structure in both aquatic and terrestrial ecosystems).
223	13-14	It will be impossible to develop “nationwide weathering rates”—since these rates show a high amount of spatial variation as a function of surficial geology and other factors such as climate.
224	8-12	The development of more comparisons with model results with observational data would greatly strengthen this overall approach by clearly showing what the models do well and where there are problems that will limit the ability to set secondary standards.

## Mr. Rich Poirot

### Chapter 4: Addressing the Adequacy of the Current Standards

**7. What are the views of the Panel on staff's assessment of the adequacy of the form of the existing NO<sub>x</sub> and SO<sub>x</sub> secondary standards? To what extent does the Panel agree with staff's assessment of the protection provided by existing standards, given the current levels, forms, averaging times, and indicators?**

Chapter 4 presents clear and convincing arguments questioning the ecological relevance the current SO<sub>2</sub> and NO<sub>2</sub> secondary standards (and many of their individual components) and concludes that current standards do not adequately protect against adverse environmental effects from SO<sub>x</sub> and NO<sub>x</sub> pollutants. Well-documented observations of chemical and ecological effects of S + N acidification in aquatic chemistry and effects monitoring programs like TIME, LTM and EMEP, combined with the observations that there are no exceedances of current NO<sub>2</sub> or SO<sub>2</sub> standards in areas experiencing effects of aquatic or terrestrial acidification or nutrient enrichment provide a general indication that current standards are not adequate. As the environmental effects from SO<sub>x</sub> and NO<sub>x</sub> occur primarily through the long-term cumulative deposition of multiple S and N compounds, combined together rather than individually, the chapter persuasively shows how the elements of the current secondary standards are inappropriate in terms of indicators, averaging times, levels and forms, as well as in their single pollutant approaches to multi-pollutant problems.

**8. What are the views of the Panel on the time frame of ecological response related to current deposition? The adequacy evaluation relies on recent NO<sub>x</sub>, SO<sub>x</sub>, deposition, and on long-term steady state ANC. Does the panel agree that long-term steady state ANC is the most appropriate representation of ANC for evaluating the adequacy of the current standards?**

Other panel members will have more informed opinions on this question. Of necessity, a NAAQS needs to be based on current air quality (and in this case associated deposition), but current effects reflect the cumulative influences of both current and historical deposition, just as future chemical indicators like ANC and ecological effects may show a substantial lag time before "recovery", and may never recover to pre-industrial conditions. I don't know of a better or more responsive indicator than steady state ANC, and think it is a reasonably strong indicator of aquatic effects which has also been observed to be responsive to increasing and decreasing S and N emissions, air quality and deposition over time.

It seems possible that some aquatic systems may be especially susceptible to effects from episodic acidification in ways that may not be directly reflected by long-term steady state ANC. I'm not sure how these kinds of systems could be better protected, but again think that other panelists can offer better advice – if this is even a significant concern.

**9. To what extent are the characterizations of ambient air quality and deposition**

**appropriately characterized, relevant to the review of the secondary NO<sub>x</sub> and SO<sub>x</sub> NAAQS, and clearly communicated?**

The characterizations of air quality and deposition presented in Chapter 4 are, for the most part, appropriately characterized and relevant to the review of secondary NO<sub>x</sub> and SO<sub>x</sub> standards. While the chapter is nominally focused on evaluating the adequacy of current standards, it also includes considerations of the potential to use current monitoring networks (supplemented by additional measurements and/or by CMAQ model results) for the development of potential new standards and/or determining compliance with them. While most of the map information is useful and clearly presented, several of the maps appear to have inaccurate legends or captions (Figures 4-8 through 4-11; see more detailed comments below). It would also be useful to include maps of total NH<sub>x</sub> N deposition, as well as the fraction of total N deposition contributed by reduced (or oxidized) N.

There are some important maps showing the relationships between atmospheric concentrations and total deposition of S and N ( $V_{S/N}$ ) presented with poor clarity in Chapter 5 (Figure 5-5). These kind of maps and the relationships they display are important and could be introduced (much more clearly), and expanded upon in Chapter 4. Presumably, it could be clearly demonstrated, through use of CMAQ ratio maps and scatter plots based on grid point comparisons, that NO<sub>y</sub> is a much better predictor or indicator of total oxidized N (NO<sub>x</sub>) deposition than NO<sub>2</sub> is, and this would directly support your evaluation of adequacy. If time/resources allow, it could also be informative to consider if (NO<sub>y</sub> minus NO) might actually be a better indicator than total NO<sub>y</sub> of total NO<sub>x</sub> deposition, since NO (is measured along with NO<sub>y</sub> but) does not deposit efficiently and may not be as good an indicator of local N deposition as are other NO<sub>y</sub> components). At some point it may also be useful to ask if NO<sub>y</sub> is a substantially better indicator of NO<sub>x</sub> deposition (in rural/remote areas affected by acid deposition) than is the sum of HNO<sub>3</sub> + PNO<sub>3</sub>. The reason I suggest asking this is that if HNO<sub>3</sub> + PNO<sub>3</sub> is an adequate indicator, then monitoring for new standards (including SO<sub>2</sub> + PSO<sub>4</sub>) could be accomplished by relatively modest enhancements to existing filter pack networks – less interesting but much less costly than adding new continuous SO<sub>2</sub> and NO<sub>y</sub> monitors at many rural/remote locations.

**Other Comments on Chapter 4**

P 101, line 7: Change “sensitivity” to “sensitive”.

P 104, lines 1 & 5: You refer to “the last review” of secondary SO<sub>2</sub> NAAQS, and then cite an EPA 1982 reference. Wasn’t there a review – or at least a decision not to revise – made in 1988?

P 105, line 12: You could add “directly” between “not” and “address”

P 105, line 19: Change “grouping” to “groupings” or “groups”.

P 106, lines 1-4: this seems like somewhat of an odd question since it is really more the effects-based monitoring programs like TIME and LTM that clearly show adverse effects occurring despite widespread attainment of the obviously inadequate current NAAQS. The subsequent

review of existing air quality and deposition networks, while helpful, seems to relate more to how well those networks could support the new standards under consideration. Toward that end, I think it might be useful to consider the extent to which existing networks might be adequate/inadequate or enhanced – both at minimal and at more scientifically ideal levels to support new standards. For example, CASTNET already measures  $\text{SO}_2$  &  $\text{SO}_4^{2-}$  and IMPROVE could be similarly enhanced by adding back-up treated filters to catch  $\text{SO}_2$ . CASTNET already measures  $\text{HNO}_3$  and  $\text{PNO}_3^-$  (&  $\text{PNH}_4^+$ ) by filter pack. Similar filter-based measurements might be added to IMPROVE, and possibly  $\text{NH}_3$  by analyzing denuder extracts. If the sum of  $\text{HNO}_3$  and  $\text{PNO}_3^-$  could be related to total  $\text{NO}_x$  deposition nearly as well as  $\text{NO}_y$  can, it (combined with the use of filter pack  $\text{SO}_2$ ) could save a lot of \$ compared to the alternative of adding new continuous  $\text{NO}_y$  and  $\text{SO}_2$  monitors in rural areas.

P 106, line 13: Does this mean that when you refer to  $\text{NO}_y$ , you mean “all oxidized N expressed as if it were NO” or do you mean “expressed as total N”. Similarly on lines 19, 20, I assume you mean that TS refers only to the sulfur from and not the actual sum of  $\text{SO}_2$  &  $\text{SO}_4^{2-}$ .

P 108, line 6: Capital V in Visual.

P 109, line 6: Change “longe term” to “long-term”

P 111, lines 12-15: I don’t get your point. You could have all the new  $\text{SO}_x$  and  $\text{NO}_y$  monitors in the world and it would tell you absolutely nothing about whether S+N deposition occurring despite attaining the current  $\text{SO}_2$  and  $\text{NO}_2$  standards is causing adverse effects. Your existing  $\text{SO}_2$  and  $\text{NO}_2$  network is adequate to tell whether standards are attained (they are) and your existing aquatic effects networks (TIME, LTM, etc.) are adequate to tell you if sensitive aquatic resources are protected (they’re not).

P 111, lines 28-29: I don’t dispute that NADP + CASTNET are valuable networks and might be useful starting points for new measurements to assess a new standard (or evaluate its effectiveness). However, it should be noted that relatively few NADP sites and no CASTNET sites are operated by state or local air agencies – whose jurisdictions might fall into non-attainment as a result as measurements made by EPA contractors (or academic research groups in the case of NADP) using funds that would otherwise be allocated to the states. Could make the states cranky...

P 111, line 30 & continuing: It’s also likely that the current configuration of NADP sites is more suitable for evaluating acidification effects than is the current configuration of (predominantly urban) air monitoring sites. So its more likely that air quality measurements might be added to NADP. However, as a practical matter, you won’t actually need NADP data to evaluate compliance with a new NAAQS, though it will be useful to evaluate whether your assumed deposition to air quality ratios are, and are remaining, reasonable.

P 112 lines 9-16: This whole paragraph needs a re-write. Also (lines 11-12) it seems a bit harsh and certainly premature to condemn CASTNET as a flimsy research effort that will never attempt to develop better operational methods. Meanwhile in other sections of the PA (like the

preceding page) you propose CASTNET as the logical starting point for a new compliance network.

P 113, line10: You could add NH<sub>3</sub> to this list of important gasses not measured by CASTNET.

P 114, line 15: Delete either “treating” or “simulating”.

P 115, line19: the semicolon should be a comma (or be consistent and also use “;” in the next line after NO<sub>x</sub>).

P 115, lines 26 & 27: Change to either “... where the more aged air masses consist...” or “with the more aged air masses consisting...”

P 115, lines 29-30: Try to be more careful and consistent with subscripts, superscripts, charge signs for ions, etc. – lest the reader waste time trying to figure out if you mean something different when you use S<sub>T</sub> and S<sub>T</sub>.

P 116, lines 10-12: Just curious, but I wonder if these runs incorporate a recently developed CMAQ bi-directional flux algorithm for NH<sub>3</sub> (seen in recent talk by Robin Dennis) that roughly doubled the effective transport distance for NH<sub>3</sub>?

p. 116, lines 18-21: I don't really buy this logic. If the (annual) NO<sub>2</sub> standard were set low enough (and attained), the deposition of oxidized N and its effects would certainly be diminished. There are better indicators than NO<sub>2</sub>, but if you used NO<sub>Y</sub>, set the level appropriately and attained it, levels of NO<sub>2</sub> will be much lower than they are now and much, much lower than the level of the current NO<sub>2</sub> secondary standard. Further, efforts to reduce NO<sub>2</sub> concentrations have not uniquely focused on reducing “NO<sub>2</sub> emissions”, but more typically emissions reductions (or permit conditions) are based on reductions of NO or NO<sub>X</sub> emissions. I like the inclusion of the Figure 4-19 NO<sub>2</sub>/NO<sub>Y</sub> ratios (inversed), but this alone isn't really sufficient to show the inadequacy of NO<sub>2</sub>. What you would really need for that would be a comparison of the superior effectiveness of NO<sub>Y</sub> as a surrogate for N<sub>OX</sub> (oxidized N) deposition. You could do this by showing CMAQ Ndep/ NO<sub>2</sub> ratio maps, similar to the Ndep/NO<sub>Y</sub> maps in Figure 5-5 (but make them legible). A test of the strength of the predictors could be shown as a scatter plot & correlation of the deposition vs. atmospheric predictors, where each point would be a CMAQ grid cell location. In theory, NO<sub>Y</sub> should be a better predictor – and especially if you constrained the comparison to rural locations where acidification may be a problem.

P 117, Figure 4-5: would it make any sense to express this in μg/m<sup>3</sup> of N, for a more direct comparison with its S counterpart (Figure 4-9), and to provide more common units and comparability in deposition/air quality (V<sub>S/N</sub>) ratio maps (Figure 5-5)?

P 120: In figure caption, “ammonia” should be “ammonium”.

PP 120-123: I think there's a problem (probably with the units) in some of these, as the scale for total SO<sub>X</sub> or ST, expressed “as μg/m<sup>3</sup> S” in Figure 4-9 goes from 0 to 10, but the scale for SO<sub>2</sub>-only in Figure 4-10, also indicated as being expressed “as μg/m<sup>3</sup> S”, goes from 0 to 20. The SO<sub>4</sub>

plotted in Figure 4-11 also looks too high to be in the indicated units of  $\mu\text{g}/\text{m}^3$  S, and so I suspect that 4-10 and 4-11 are probably showing  $\mu\text{g}/\text{m}^3$  of  $\text{SO}_2$  and  $\text{SO}_4$  respectively (not just the S).

P 124: The VIEWS figures are useful, but you might include a footnote explaining that the “plume” in the central US is an artifactual error from the interpolation & plotting routine, and not “real”.

P 130: The scale indicates “ppbv” but the indicated ratio that’s plotted should be unitless.

P 131, line 5: Another good example of the confusing use of terms. In this case, I assume, but don’t really know, that you’re using “ $\text{NO}_x$ ” to mean “the nitrogen from the total wet and dry deposition of all oxidized nitrogen compounds”, and that by “total atmospheric deposition” you mean the “total wet and dry deposition of all reactive nitrogen”. This reminds me that it would have been useful to include maps of total  $\text{NH}_x$  N deposition, and of the ratio of total N deposited by oxidized N compounds.

p. 131, line 24: It’s not clear where “summarized above” is pointing to. If you mean way back in section 1.4, why not say so, and save the reader a lot of unnecessary page flipping.

P 134, lines 21-26: Could you also include % of lakes exceeding CL for ANC of  $100 \mu\text{eq}/\text{l}$ , since that threshold is still on the table as a possible indicator level. Isn’t it?

P 135 lines 2, 3 and elsewhere: You frequently invoke “brook trout” as a “sensitive species”, but I tend to think it as one of the more acid-tolerant species (Charlie or Myron can clarify this). Also, it’s not clear in this sentence whether an ANC of 20 is a base level which could dip even lower during an extreme event, or the level to which some baseline higher ANC might drop during an extreme event. I think this is an important concept that could be addressed in more detail. Finally, do you really mean to indicate a population decline during an event (fish kill)?

P 136, line 28 and lots of other places: Use either BC/Al or Bc/Al but not both.

P 141, line 28: Am I thinking backwards or shouldn’t “sensitive ecosystems” in this context be ones where “N is limiting”, such that excess N would cause shifts in species?

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

**10. What are the views of the Panel on staff’s proposed conceptual framework for the structure of a multipollutant, ecologically relevant standard for  $\text{NO}_x$  and  $\text{SO}_x$ ? 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  $\text{NO}_x$  and  $\text{SO}_x$ ?**

The proposed conceptual framework and the structure of the proposed standard do an excellent job (under very challenging Clean Air Act constraints) of representing the complex linkages between ecological effects, aquatic chemistry, atmospheric deposition and air quality. It is inconvenient that a direct measure of ANC in surface waters, or measurements of deposition of S

and (total reactive) N couldn't be used as a more direct basis for a NAAQS. However, given the limitations of the current CAA, the proposed structure employs some clever and innovative mechanisms to link air concentrations to ecological effects, recognize the combined influence of S and N, accommodate the varying inherent sensitivities of different ecosystems, and incorporate – without directly regulating - the additional influence of reduced N.

**11. What are the views of the Panel on the relevance of the conceptual design for developing standards to protect against aquatic acidification effects?**

**a) What are the views of the Panel on staff's proposed options for the ecosystem acidification model to represent the ecological response function?**

Others on the panel will have much more informed opinions on this than I could offer. On a related topic, I'm not sure that the concept of episodic acidification is adequately addressed in the acidification model(s) presented chapter – or elsewhere. It seems to be assumed that selection of a specific ANC limit – of say 50  $\mu\text{eq/l}$  – would be intended to protect against both chronic and episodic effects. If this is the case, it should be stated explicitly. If there might be exceptions, for certain types of catchments or ecosystems, these should also be presented along with indications of how selection of alternative ANC limits might guard against these effects.

**b) What are the views of the Panel on the relative merits of the two techniques for calculating Neco, the parameter representing the amount of deposited nitrogen that is available for acidification due to uptake, denitrification and immobilization?**

Others on the panel will have much more informed opinions on this than I could offer. From the text explanation on p. 178, it would seem like equation 3 is both simpler and more readily accessible or “testable” through direct methods than equation 2. I would think it would be applicable, however, only in situations where (or when) “N breakthrough” has already occurred and is directly measurable, and therefore would only reflect current retention rates, which well may decline further with continuing deposition. Equation 2 may be a better, more protective approach to guard against longer-term future effects and assure that current rates of N deposition are “sustainable”.

**c) What are the views of the Panel on staff's proposed options for developing acid sensitivity classes to categorize the national landscape? Is it appropriate to base this classification on bedrock geology? Should multiple criteria be used to inform the sensitivity categories?**

This sounds like a reasonable approach, and possibly other variables (surface soil composition and depth, weathering rate, slope, elevation, etc. could also be useful. It's difficult to judge this without seeing an example, but in general, I think it could be an extremely valuable exercise to go through in some detail, regardless of what the eventual end use(s) is (are). Conceivably multiple approaches to developing sensitivity classes could be considered and then evaluated – by comparing measured vs. predicted ANC using the different sensitivity classes. Would it be possible to use relatively abundant surface water chemistry data to guide this process?

**d) What are the views of the Panel on staff's proposed options for how to aggregate acidification modeling from the catchment-scale to represent acid-sensitivity categories at the national-scale?**

It's difficult to evaluate these options in the abstract, without seeing comparative examples. The approach of specifying a percentage of water bodies within a given area sounds reasonable, but would be critically dependent on how the areas were selected. Selecting a critical % of lakes to protect seems like a somewhat arbitrary decision in the first place, and when combined with optional methods of selecting the (size of, and distribution of lake characteristics within) spatial areas, the decision process could be extremely arbitrary. Within a small area at high elevation on the western slopes of the Adirondacks, 100% of lakes will exceed their critical loads to maintain a protective ANC, but as the area is expanded in size, the % of acidified lakes will eventually fall to single digits if aggregated at the statewide level, so where do you draw the line?

I wonder if it would be possible to modify the geographical area concept to include only the population of surface waters within that area to include only those considered "potentially susceptible" to acidification – based on characteristics of their underlying bedrock, soils, etc. This would minimize the importance of the specific areal selection, since the % affected metric would apply only to the lakes considered susceptible in the first place. Or maybe model estimates of preindustrial ANC levels could be used as an index of inherent sensitivity and a decision of adverse affects could be based on shifts in ANC categories over time, or based on a limit to the % change in ANC from preindustrial conditions within the population of potentially susceptible lakes in a given area.

In areas where acidified streams are at issue, it would be necessary to have some metrics for selecting the appropriate sections of streams to be evaluated, and this doesn't seem to be discussed in much detail in the Policy Assessment. As with the affected lakes, some metric that reflects past ANC changes over time might be a useful way to judge adversity of current conditions. Conversely, it might be useful to consider a progress-based metric (% increase in ANC, or % shift between ANC categories over time) to determine future compliance with the NAAQS. I think there are other examples where one test is applied to get an area into non-attainment, but a different test is required to get out of non-attainment.

**e) What are the views of the Panel on staff's suggested method to account for reduced nitrogen in the deposition metric?**

It's unfortunate that reduced N can't be more directly recognized as an important, unregulated pollutant and steps taken to reduce its emissions to the atmosphere and surface waters. However, given the constraints in the CAA definition of nitrogen oxides, staff have devised a rather ingenious method of accommodating its influence without regulating it directly. I would assume, however, that if  $\text{NH}_x$  deposition rates were reduced in an acid sensitive area, then proportionately higher levels of  $\text{S} + \text{N}_{\text{OX}}$  deposition could be accommodated.

**12. What are the views of the Panel on the relevance of the conceptual design for developing standards to protect against terrestrial acidification effects? Terrestrial nutrient enrichment effects? Aquatic nutrient enrichment effects? Does the panel have**

### **suggestions on additional data or methods that might enable EPA to expand the current aquatic acidification approach to cover additional effects?**

The conceptual design for protection against terrestrial acidification effects seems reasonable and consistent with recent advances in the science and assessment methods. The BC/Al ratio seems like an excellent “chemical indicator” (directly measurable and predictable via models), which is directly responsive to deposition changes on one end and directly predictive of ecological effects on the other end. The currently considered range of levels (0.6 to 10) seems rather broad (relative to ANC ranges of 50 to 100, although from the comparisons of aquatic and terrestrial sensitivities provided, would seem to suggest that a BC/Al of 10 at the most protective upper limit considered is most similar (similarly protective) to ANC at 50  $\mu\text{eq/l}$  (least protective) lower limit. In a general way, it would seem logical to expect that if chronic long-term deposition of acids, passing through soils supporting terrestrial ecosystems enter aquatic systems sufficiently un-buffered by associated cations or enriched in toxic Al to cause adverse aquatic effects, then we should expect that there would also be long-term terrestrial effects in those upstream watershed areas. That is, we should expect that ideal aquatic and terrestrial effects indicators might show similar sensitivities.

Your approach for aquatic effects is more completely developed, and you could probably justify a position that it is logical to start simply with a single indicator to protect against aquatic acidification – which would in many cases also afford added protection against terrestrial acidification effects. However, I think you could also make a good argument that an approach using both ANC and BC/Al indicators (& associated deposition & AQ limits) would provide better protection and over somewhat broader areas as there may be sensitive soils within catchments that have more well-buffered soils in other areas such that there are terrestrial effects upland of lakes or streams which are relatively insensitive.

Confidence is lower for setting specific standards for terrestrial and aquatic nutrient enrichment in the present review cycle, and a focus on acidification could be justified. I think however that the consideration of the carefully derived loading limits – and approaches to apportioning and reducing them – that have been / are being developed through EPA’s TMDL process might be given more consideration in the future. There might be some innovative ways of combining TMDLs and NAAQS (for both acidification and for nutrient enrichment effects). In seeking reductions from point source or direct runoff discharges, some TMDL exercises (CT River discharge to Long Island Sound, for example) are assuming specific future CAA reductions in atmospheric N deposition loadings as part of their basis to specify and allocate reductions in direct N discharges. Conceivably a N deposition-related NAAQS could be considered exceeded throughout a watershed where downstream TMDLs are exceeded, and in the implementation phase, costs & ancillary benefits of decreasing N emissions from various source categories could be considered jointly. It should also be noted that TMDLs for critically acidified lakes have been developed in some states (VT & NH – based I think on ANC limits of 50 & 60  $\mu\text{eq/l}$  respectively) and there may be some logic to combining the NAAQS & TMDL processes – as if there were a single EPA...

### **13. What are the views of the Panel on the critical uncertainties associated with the conceptual design of an ecologically relevant multi-pollutant standard that need to be**

## **characterized in terms of their potential implications for the secondary standards?**

Others on the panel will have much more informed opinions on this than I could offer. I think each of the acidification models and the CMAQ model have been subjected to periodic performance evaluations and sensitivity analyses, and that some of the most critical (most uncertain) model inputs, processes and parameters have been identified. Associated efforts are also periodically undertaken to identify and strengthen the weakest links in these models (or to develop better new ones) such that we can expect model-associated uncertainties to improve in the future. A strength of the proposed approach is that the AAPI is in effect a estimate derived from atmospheric measurements which has an approximate, measureable aquatic counterpart (ANC) indicator (and there are also various intermediate modeled deposition metrics that can be evaluated with measurements). As such, the net effect of the cumulative uncertainties and biases of the proposed regulatory metrics can be continually evaluated and refined.

## **14. To what extent do the figures and examples aid in clarifying the text? Should more or less information of this type be included in the second draft?**

Given the complexity of the subject matter, the many associated equations and difficulty (for me at least) of “seeing” many of these concepts in the abstract, I found all of the illustrations and example calculations (from actual data) very helpful and would like to see more of the same. One figure in particular that seems important and could be presented more clearly is the display of modeled VS/N ratios (barely legible) in Figure 5-5. Larger-scale versions of these plots would help, along with scales selected to better show the ranges of values. Possibly use of the same scales for both the S and N ratios could be informative. It could also help to show zoomed-in versions the cover identified sensitive areas in the Adirondacks, Shenandoah, etc. It strikes me also that the accuracy of these CMAQ estimates could be tested (easily for S, not so easily for N) by comparing the modeled ratios to those from CASTNET & NADP measurements. You presented some CMAQ model performance evaluation information previously, but when model results are ratio-ed, there’s a potential for errors/biases to get compounded (or offset)

I had a difficult time making it through this chapter with confidence that I was understanding the details of all the points being presented (in the 23 separate equations). I think this was due partly to it being outside my expertise, partly to the introduction of many new unfamiliar variables, and partly due to the inherent complexity of the subject (further confounded by the CAA constraints). However, I also think that the information could be presented more clearly, and here are some suggested approaches toward making this no more confusing than it needs to be:

- Be precise and consistent in your terminology (and triple check for typos especially in the equations). I see no useful reason whatsoever to use the terms  $\text{NO}_x$  and  $\text{NO}_y$  to mean the same thing.  $\text{NO}_x$  has inherent meaning (sum of NO and  $\text{NO}_2$ , including the oxygen) to the air quality community and you can’t revise this meaning with a disclaimer up front that “when we say night we really mean day”. This gets further confused by the occasional use of the term  $\text{N}_{\text{OX}}$  to mean oxidized nitrogen”. I actually like this term (we can pronounce it “ennox”) and think it could be used nearly interchangeably with  $\text{NO}_y$ , with the possible distinction that the former relates to the concept and the latter relates to our attempts to measure it. But I also see an occasional  $\text{N}_{\text{OY}}$  – which I assume is a typo (pronounced as “annoy”), but then that makes me wonder if/when  $\text{NO}_x$  is used as

intended. Is  $g(\cdot)$  the same as  $g$  the same as  $G$ ? If you mean  $\text{SO}_4^-$ , write it that way and don't use  $\text{Cl}$  when you mean  $\text{Cl}^-$ . Use  $\text{BC/Al}$  or  $\text{Bc/Al}$ , but not both, etc.

- Number the equations sequentially from the beginning of the document – or at least from the beginning of each chapter (rather than anew each subchapter) so that there won't be 3 different equations #4 in a single chapter.
- For each chapter, or at least for chapter 5, or maybe as a stand-alone appendix applying to all chapters: develop a glossary-like list or table of all terms used in your equations, along with definitions and other pertinent information – possibly including things like how the term would be measured or estimated, its units, etc. The Table A-3 list of variables used in the SSWC model presented in Appendix A is a good example of what I mean.

### Other comments on Chapter 5

P 145 & 6: The text describing Figure 5-1 refers to squares, triangles and circles, but the figure contains only rectangles, diamonds and ovals.

P 148, lines 9-12: Its difficult to grasp this concept in the abstract. Could you give an example? Are you thinking about 5 bins or 25?

P 148, lines 16-18: It might help to have a diagram to illustrate this concept.

P 150, line 21: Add “of” after “deposition”.

P 153, lines 29, 30: Try to be consistent with use of charge signs for ions.

P 167, line 10 (& elsewhere): Be consistent in your use of terms:  $\text{BC/Al}$  or  $\text{Bc/ Al}$ ?

P 167, lines 26-30: This summary of aquatic vs. terrestrial CL sensitivity seems inconsistent with the similar summary on p ES-13 of the executive summary.

P 168, lines 1 & 2: Is this observation that for the Shenandoah, the aquatic CLs provided much greater protection than terrestrial true for  $\text{BC/Al}$  ratios of both 10 and 1.2?

P 169, line 3: Add something like “addressing” between “for” and “these”.

P 171 line 2: aggregated deposition.

P 172, line 25 “Table 1” should be changed to “Table 5-3”.

P 172, line 28: The importance of large particle S & N deposition may be overstated here – especially with respect to acidification effects, which will tend to occur at higher elevation locations remote from coarse particle sources (which don't transport very far). Also, S or N in coarse particles is often in association with crustal material (containing  $\text{Ca}^{++}$ ,  $\text{Mg}^{++}$   $\text{K}^+$ ) or  $\text{Na}^+$  from sea salt – hence carrying its own buffers. At CASTNet sites (where open-faced particle samplers presumably include coarse particles), all particles contribute less than 6 % of total S deposition at sites with  $\text{S dep} > 5 \text{ Kg/Ha/yr}$ . For CASTNet nitrate deposition, total particle nitrate deposition averages  $< 2\%$  of total nitrate deposition across the whole network. So its not

likely that coarse particle deposition accounts for much of the total S + N deposition load at any sites where aquatic acidification is a problem.

P 173, line 1 simulations of.

P 173, line 5: text refers to “inverse”  $V_{S/N}$  but Figure 5-5 caption does not indicate “inverse”.

P 173, line 10: “6 for”. Also this seems like an example of something I’ve noted on several occasions of referring to something “deposition load tradeoff curves” that hasn’t been explained yet. Makes it harder to understand...

P 173, line 11: “Figure 1” should be “Figure 5.5”, I think.

P 173 line 15: “Figure 3” should be Figure 5.7.

P 174, Figure 5-5: I would like to see these at larger sizes with scales that better show the range of values. These ratios seem very important (see response to Q 14). On a somewhat related topic, I would think that of all the  $\text{NO}_Y$  components, NO is likely to have the lowest deposition velocity, and otherwise be least reflective of N deposition at any specific location. I believe that in making  $\text{NO}_Y$  measurements the sampling is typically switched through and then bypassing the converter, such that concurrent NO and  $\text{NO}_Y$  data are available. Possibly then [ $\text{NO}_Y$  minus NO] might actually be a better AQ indicator of  $\text{N}_{\text{OX}}$  deposition than total  $\text{NO}_Y$  – both in reality and in CMAQ output. I would imagine this could be tested relatively easily with CMAQ.

P 178, line 28: Is this basically the equation for “deposition load tradeoff curves” referred to previously and later in this section? If so, why not describe it as such here.

P 179, line 19: Is it really the “high variability in the data set” per se that results in “only 32%” of water bodies not exceeding their critical loads, or is it more that the selected population of lakes includes many which are relatively well buffered? Or are you making a point that use of “mean  $\text{DL}_{\% \text{ECO}}$ ” can be problematic for areas with high variability? This isn’t clear to me, but I’m concerned that use of a metric based on % lakes in “an area” becomes an important, but arbitrary factor (based entirely on how the areas are spatially defined).

P 180, 2<sup>nd</sup> line of Table 5-4 caption note: Delete either “across” or “on”.

P 180, Table 5-4: From the descriptions of equations 2 and 3 on p 178, I would have assumed that for a given catchment either one equation or the other would apply (# 2 projecting the long-term additional N a system could handle before leaching – or # 3 based on measurements where saturation has already been reached). So I’m surprised to see in Table 5-4 that there are summary statistics for  $\text{N}_{\text{eco}}$  presented using both equations. I’m also surprised that the results for both  $\text{N}_{\text{eco}}$  and  $\text{DL}_{\% \text{ECO}}$  are so different depending on which equation was used. Can this be explained more clearly?

P 184, lines 1, 2: Eight significant digits for these “aggregate effective deposition velocities” seems a bit much. I also agree with your plans to change the term to something like “deposition

ratio” for clarity. Why not do it the other way around and call it a deposition ratio in the text and use your footnote to explain that you used to use another, less appropriate term.

p. 187, line 9: You indicate here that equation 5 describes the “atmospheric concentration tradeoff curves”. Is this correct or should it be equation 6?

P 187, line 2 & elsewhere: I think the term  $N_{OX}$  is a logical way to describe “oxidized nitrogen”, but think it might be helpful to add a footnote caution that this should not be confused with  $NO_X$  (which you have already confused with  $NO_Y$  sometimes but not always). Also, I note that in equation 11, p 186 you use the new term  $N_{OY}$  which I assume is the same as  $NO_Y$ ?

P 187, lines 11, 22: The term “ $g(\cdot)$ ” in equation 11 has become just “g” in equation 12 (then back to “ $g(\cdot)$ ” in equation 13). Do these changes have a meaning (that I don’t understand because I don’t know what  $(\cdot)$  means)? And are either of these the same thing as “G” referred to on p 198, line 23 and elsewhere in the PA?

## Mr. Dave Shaw

### General Comments

I believe a strong case has been made throughout the REA, ISA and this policy assessment (PA) that the current SO<sub>x</sub> and NO<sub>x</sub> NAAQS are not adequate or appropriately protective of welfare. A multi-pollutant strategy is appropriate and feasible to propose at this time with some considerations. As I have mentioned in previous comments, there are not enough monitors in most areas of the country to adequately characterize the sensitivities of regional ecosystems. While I do feel it is appropriate to use the existing data, and depend heavily on modeling at this time, it is necessary to commit to building the existing monitoring network throughout the country to better develop this NAAQS. Also, the models will be most useful if they are shared with all states which may wish to assist with refining them.

On that same note, I suggest a commitment of some kind be developed within a year which is specific about providing adequate data to quantify inputs and results. A plan should be outlined so that at the very least we can identify where monitoring is needed and perhaps prioritize to make the best use of limited resources. This multi-pollutant approach will help in decreasing resource needs by having, for example, one monitoring location for several pollutants. This proposed NAAQS is innovative but complicated and very model dependent, and because of that, it calls for these other commitments. As I have previously indicated, we should be aiming to significantly improve our ability to make modifications to the next NAAQS (which hopefully will be quicker than the last) based on significantly improved data and model outcomes. The best scenario would be that these monitors would already be in place. Our atmospheric conditions and pollutant loads are changing; we don't have any extra time for delaying.

### *Multi-Pollutant Approach*

Multi-pollutant air quality planning is currently being evaluated in New York State. This may be a more efficient way to run and plan a State Air Quality program [than one pollutant at a time]. The ability to plan and regulate multi pollutants with one policy should be less resource intensive as well as give us a better understanding of co-benefits between pollutants. So far, we have found that multi-pollutant planning tools and approaches lend themselves well to supporting implementation and enforcement of multi-pollutant secondary standards such as NO<sub>x</sub> and SO<sub>x</sub>. Since our endpoints, such as surface water Acid Neutralizing Capacity (ANC), are far removed from the emissions point sources, a multi-pollutant planning approach can be used to model the policy drivers and the resulting wide range and variety of benefits.

I believe that a multi-pollutant planning approach that integrates air quality and climate goals may prove to be the best action to take to meet the forthcoming multitude of federal and state environmental requirements. NYSDEC is in the process of developing a comprehensive air quality management plan (AQMP) that is multi-pollutant in nature with the intent that it provide for a more efficient and proactive pollution control process.

### *Flexibility*

The PA indicates that future drafts will be expanded to include effects other than aquatic acidification which I support as well as future actions in this direction. This assessment is based on available data and modeling and at this time, there is adequate data from New York's Adirondack Region as well as the Shenandoah Region concerning surface water acidification. Other regions in the country that currently do not have sufficient monitoring data or modeling efforts to characterize their own sensitive ecosystems may find that they must be more concerned with terrestrial acidification, nitrification or fresh water/coastal eutrophication. Therefore, I suggest that sufficient flexibility be built into the policy to allow for these future monitoring/modeling efforts and characterization.

### *Models & Data*

Appendix A provides a good overview of the models, but could be improved by including an assessment of reliability and a description of what range of error is acceptable. It would be helpful to know what data exists for how many watersheds for what range of time. Page 28 mentions that there is a lot of variability from year to year; it would be helpful to know what is considered "a lot." Also, it would be helpful to state how many years of data or monitoring sites would raise the reliability of this data by 2 times, 10 times, etc.

I am concerned about the availability of accurate weathering data since this is such a critical variable in the models. Have the lacking weathering rates been developed for the sensitive regions?

Appendix A also discusses the supporting datasets for the MAGIC model. Datasets and the models should be made available to not only the EPA, but also the States which are responsible for implementing the NAAQS which are being proposed. I want to be certain that whatever models will be utilized, that the States have access to make refinements. This should help advance EPA's progress on the same.

### *Reliance on ANC*

ANC is an available, good indicator; however, it might be misleading to call it an "excellent" indicator (p. 134, line 16). There are other indicators that could be discussed and utilized, for example, when toxic inorganic monomeric aluminum ( $Al_{im}$ ) is not present, but ANC is low, then organically derived low pH and ANC might be more of a driver than mineral acidity from an air quality problem.

While ANC levels may be current (lines 28-29), the biological data associated with them are not. The ANC and fish diversity relationship is based on a 1469 lake data set from 1984-1987, and the losses in health and reproductive capacity from other sources may be just as old or older. While evaluating monitoring data, it would be helpful to include updated surveys in the modeled lakes.

### *Bedrock geology as an indicator*

On pages 162 – 164, the discussion of acid sensitivity based on bedrock and multiple landscape features should also include a discussion of glaciated versus non-glaciated landscapes. In the glaciated Adirondack region, there are many areas where soils do not reflect the underlying bedrock. Surficial geology and depth of soil would be useful to discuss in this section.

### *Critical loads*

I would like to see more discussion on critical loads data and models. While I understand that we would like to use landscape and effects data which are specific to the United States, I believe Canada and Europe have a wealth of experience in the use of critical loads models and their successes and limitations. From a policy perspective, it would be worthwhile to analyze their use of critical loads and what they have learned from various model approaches. Those experiences could inform the US NAAQS review and process.

## **Charge Questions**

### **Chapter 9**

**25. What are the views of the Panel on the preliminary staff conclusions regarding the adequacy of the current standards, the need for an integrated multi-pollutant structure for revised standards, and the proposed form of the joint NO<sub>x</sub> and SO<sub>x</sub> standards for aquatic acidification?**

**26. What are the views of the Panel on the overall characterization of uncertainty as it relates to the determination of an ecologically-relevant multi-pollutant standard for NO<sub>x</sub> and SO<sub>x</sub>?**

The document clearly demonstrates evidence that the current standard is not adequate due to the fact that ecosystems are still impacted. The short term average NO<sub>x</sub> and SO<sub>x</sub> concentrations are not protective or appropriate so there is a need to consider a new strategy. The evidence in the documents show that this is a multi-pollutant problem so a multi-pollutant approach is appropriate.

The current approach is acceptable because it is the most commonly used and understood. Knowing this, it would be best to stay mindful of the limitations of ANC as an aquatic endpoint and develop the policy to be open to refinement including other measures for aquatic endpoints such as Al<sub>im</sub> and Base Cation Surplus (BCS).

That being said, I must re-state that we must make additional monitoring available for locations which currently have no monitoring available. While this approach is the best with what is available, we may find with additional data collection, that different ecosystems require different kinds of protection.

## Specific

### *Draft Executive Summary (ES)*

ES-3 para 4. Line 4 delete 'acid'.

“ para 4, line 6 substitute 'salmonids' or 'trout' for 'Atlantic salmon' if this is based on Adirondack evidence [Atlantic salmon are not common, found in only 13 out of 1469 lakes sampled during the ALS as opposed to brook trout found in 579 lakes].

ES-4 para 1. Since this is the first discussion of this topic it would be the best place to discuss the connection with springmelt (this occurs somewhere later in the ES). Also the ANC levels discussed are annual averages levels. Need to convey this clearly because, for instance, a springmelt ANC of 50 would not be associated with death or loss of biota, the effects description is associated with a baseflow or an annual average.

ES-5 full para 2 starting with Current acidification.

Line 3 Add the year associated with 78% Adk lakes, and 'baseflow (summer) or annual (it's one or the other, not both)' before ANC.

Line 4 Delete bluegill, since they are uncommon in the Adirondacks (occurring in only 4 out of 1469 lakes surveyed during the Adirondack lakes survey in 1984-87) and not raised to the level of recreationally significant in this region.

“ para 3 line 2 add after 'recent' the year of the study.

ES-7 para 3. Bedrock geology may not be the best predictor of acid-sensitivity in the Adirondacks due to glaciation (see comment in main PA document).

ES-9 on use of equations. It would be more useful to describe the AAPI that use an equation (not recommended for an exec sum).

“ First para under Options. Delete entire.

ES-10 para 1. An important paragraph, it should stay in but be modified to be more clear and less wordy/redundant. It might also be moved up to the first paragraph in that section.

“ para 3 line 1. Delete 'using equation 1 above'.

“ para 3 line 7 “an AAPI level” repeated.

“ para 3 last line. Run on sentence.

“ Para 4,5,6 Suggest better to explain in text than to use model terms.

ES-12 full para 2. Important point. How to treat naturally organic ecosystems is a key aspect. How does this overall approach (including how well does MAGIC treat this?) come close to adequately addressing this?

“ full para 4. Suggest delete, since all was stated earlier in the ES.

ES-13 last para first line after last review, include date.  
last para sentence beginning with “In addition..” delete “substantial” and “information, based on”  
and “rigorous” because this could unintentionally mislead to belief that data and modeling are  
abundant.

ES-14 bullet 1 line 1 Some language is missing.  
“ bullet 3 end of sentence, add “among others.”  
“ bullet 6 Clarify replacing the current forms of what?

*Policy Assessment*

Pg 219 lines 9-10. Add after studies “since 1991-93”.

Pg 221 line 6. Is it “strongly” supported?

## Dr. Kathleen Weathers

Overarching comments:

I think this first Policy Assessment for the Review of the Secondary National Ambient Air Quality Standards for NO<sub>x</sub> and SO<sub>x</sub> is an excellent start. The EPA staff should be commended.

Some general comments

Make sure to label all figures with scale and direction, and provide clear figure legends. Also be careful not to report too many (i.e., not significant) figures (e.g. Figure 2-9, deposition of 66.507??).

References are uneven and inconsistent.

For all the analyses, especially the components of the AAPI equation a spatial scale must be decided upon. While watersheds are ecologically useful for mass balance calculations, it may be that geology, or even more likely, the spatial resolution of the deposition estimates may control the scale at which analyses can be performed. I suggest laying out the logic, issues and constraints of choosing spatial units for analysis.

Relevant to Chapter 2 and 3, especially: I have articulated the concern in the past, and it was brought up again during the meeting, that interpreting the relationship between fish presence and abundance and ANC in the absence of information about stocking for the region under consideration (which may be the primary control on presence and abundance for many species) is difficult to defend. At a minimum I suggest not including in the analysis (whether ecological or economic) sportfish whose abundance and presence is largely controlled by stocking.

As has been pointed out, there are “enhancing” effects of nitrogen deposition for some forest species and systems that have been documented, often in combination with a changing climate (temp, CO<sub>2</sub>) (e.g., Thomas et al. 2010 and references therein). This information should be included, perhaps as a preamble to documenting the detrimental effects of deposition/NO<sub>x</sub> and SO<sub>x</sub> (which is the focus of this document).

Please use common units throughout the document for displaying data in figures and in tables, whenever possible (e.g., kg/ha-yr vs meq/m<sup>2</sup>-yr).

Responses to Charge Questions:

### CHAPTER 6

15: Developing an AAPI linked to key determinants of aquatic ANC is an interesting and defensible approach to developing an ecologically relevant standard. And, yes, I think the secondary standard options identified by the staff can draw upon existing scientific and technical

information. Of course, the devil is in the details (including uncertainties), which is no surprise, but will require significant work (see 16).

16: There are, in fact, considerable uncertainties associated with all steps of developing an APPI, and, of course all of the modeling processes involved in every step of the analysis, from emissions to deposition to effects. It is important to identify where the major uncertainties are and how they affect bottom lines. For example, matching temporal and spatial scales in various components of the AAPI remains an issue. How should uncertainty be characterized with integrity? I note that there is much ongoing research focused on uncertainty, how to characterize it, how to communicate it (witness efforts surrounding the IPCC).

I had actually expected, after suggestions and concerns that arose out of our last CASAC meeting, and because of their importance to the policy process, that the section on uncertainties would have been developed further in this document. It will be crucial.

However, that there are many uncertainties should not make scientists or policy makers reel (or at least reel away from making decisions). It is the state of the science (it is the state of life, in fact), and what is known should not be discounted. For example, what we do know about annual wet+dry deposition of S and N across the US based on monitoring data and models is vastly more than we knew a decade ago, and it is important not to obscure that fact. That there may be 100% uncertainty is better than not having any estimate whatsoever; it is at times important to contrast uncertainty with no knowledge at all!

17: Aggregation has some similarities with using ratios (e.g. stoichiometric ratios). It collapses all kinds of complex variables into a number. It's simpler to understand (at least at first pass), but can obscure important patterns and processes, especially when aggregates change in value, sometimes as a result of component variables changing in different directions. It is therefore important to understand the sensitivity of the various components.

18: CMAQ modeling is a work in progress and highly uncertain, as has been noted before, but perhaps no more uncertain than, say, the MAGIC model. As has also been pointed out before, there is a pressing need to compare CMAQ to other ground-based estimates of deposition, such as NADP + CASTNet sites (e.g., Weathers and Lynch 2008). For example, in a preliminary analysis, while CMAQ and CASTNeT dry deposition estimates compared well, the concentrations did not, suggesting that both deposition velocities and concentrations vary between the two approaches, in opposite ways (Weathers and Lynch 2008).

It is critically important to recognize the scale over which deposition estimates can be produced and how they may, or may not, be useful in distinguishing deposition estimates for watersheds, for example, that are geographically close together. CMAQ's spatial resolution for this document is 12-36 km (page 114). That said, I think that the CMAQ model is the best model to use for the AAPI calculation for comparisons on a regional or national scale. One of its strengths is that wet+dry deposition can be estimated, using a common method, across the country, and that urban areas are included (see Pardo et al. 2010). The weaknesses include an inability to scale down to watersheds, or areas where the most intensive ecosystem response variables are measured (e.g., lakes, watersheds that span < 12-36 kms in area). All of these concerns

notwithstanding, use of CMAQ and NADP + CASTNeT (including using them to model deposition at finer spatial scales and across heterogeneous terrain, e.g. Weathers et al. 2006) are state-of-the-art, currently.

19: It has parallels with the critical loads approach used for the EU and elsewhere; that approach has been in use for awhile.

20. It depends upon the use/goal, of course. In general, I think that annual (or, in some cases, multi-year) averages are more reasonable than short-term (weeks, months, even quarters) estimates for CMAQ, as well as other modeling results if the goal is to examine annual ecological responses that have seasonal responses or shortish lag times. Experience with P-Net and many other models suggests that “getting it right” for shorter time steps is extremely difficult, and perhaps not a reasonable goal. However, interannual variation can be high, based on weather patterns and emissions, especially for wet:dry ratios; it would be useful to discuss further the rationale for 3-year averages.

21: Expansion of biological (soil, vegetation), hydrological/chemical as well as atmospheric (chemical species, such as NH<sub>x</sub>, and spatial extent over which measurements are made) monitoring needs to be significantly enhanced.

22: Other panelists are better suited to answering (technically) this question. I found the section confusing.

Overarching note: I think it essential to identify and remove from the AAPI analysis “naturally” acidified aquatic ecosystems (as a result of organic acidity vs strong mineral acidity), as was suggested in a few places throughout the document.

## **CHAPTER 7:**

23: I am intrigued by the approach, and appreciated the first-pass comparison. Aquatic and terrestrial systems behave quite differently in the biogeochemical, temporal, and spatial processing of N and S, of course. And, S and N as ions or elements have different chemical (e.g., valence state) and physical properties that influence their (contrasting) behavior within ecosystems. The standards set for aquatic protection are likely to be more conservative (i.e., stringent) than terrestrial protection because, in part, the ecological responses to deposition ramify through the terrestrial ecosystem and, to a certain extent, are expressed in the aquatic system, as pointed to in this document.

Should a terrestrial acidification standard be proposed, a key component will be how to discern natural acidification (e.g. as a result of such processes such as respiration, etc.) from anthropogenic acidification.

24: I need more clarification on the question here.

References:

- Thomas, R.Q., C.D. Canham, K.C. Weathers and C.L. Goodale. 2010. Nitrogen deposition increases regional carbon storage and shifts competitive balance among trees. *Nature Geoscience* 3: 13-17.
- Weathers, K.C., S.M. Simkin, G.M. Lovett, and S.E. Lindberg. 2006. Empirical modeling of atmospheric deposition in mountainous landscapes. *Ecological Applications* 16:1590-1607.
- Weathers, K.C. and J.A. Lynch. 2008. How Much Atmospheric Deposition Across the US: Comparison of N and S Deposition Estimates. *Eos Trans. AGU*, 89(53), Fall Meet. Suppl., Abstract: B42A-01.