



UNITED STATES ENVIRONMENTAL PROTECTION AGENCY  
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May 17, 2011

EPA-CASAC-11-005

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

Subject: CASAC Comments on the *Policy Assessment for the Review of the Secondary National Ambient Air Quality Standards for Oxides of Nitrogen and Oxides of Sulfur (February 2011)*

Dear Administrator Jackson:

The Clean Air Scientific Advisory Committee (CASAC) Oxides of Nitrogen and Sulfur Oxides Secondary NAAQS Review Panel met on February 15-16, 2011 to consider the information in EPA's Policy Assessment for the Review of the Secondary National Ambient Air Quality Standards for Oxides of Nitrogen and Oxides of Sulfur (February 2011) and to provide our recommendation on the review of the standard. The draft report was approved by the chartered CASAC at the public teleconference on May 12, 2011. Below, we offer our general comments and recommendations. The CASAC and Panel membership is listed in Enclosure A. More detailed consensus comments are presented in Enclosure B. Enclosure C is a compilation of individual panel member comments.

Overall, we reiterate our previous comments from the review of EPA's second draft of the Policy Assessment: "EPA staff has demonstrated through the Integrated Science Assessment (ISA), Risk and Exposure Characterization (REA) and the draft PA that ambient NO<sub>x</sub> and SO<sub>x</sub> can have, and are having, adverse environmental impacts. The Panel views that the current NO<sub>x</sub> and SO<sub>x</sub> secondary standards should be retained to protect against direct adverse impacts to vegetation from exposure to gas phase exposures of these two families of air pollutants. Further, the ISA, REA and draft PA demonstrate that adverse impacts to aquatic ecosystems are also occurring due to deposition of NO<sub>x</sub> and SO<sub>x</sub>. Those impacts include acidification and undesirable levels of nutrient enrichment in some aquatic ecosystems. The levels of the current NO<sub>x</sub> and SO<sub>x</sub> secondary NAAQSs are not sufficient, nor the forms of those standards

appropriate, to protect against adverse depositional effects; thus a revised NAAQS is warranted.” (Russell and Samet, December 9, 2010, EPA-CASAC-11-003)

The final Policy Assessment is substantially improved from the last draft, and EPA staff has responded to most of our concerns raised in our previous comments. The Policy Assessment develops a framework for a multi-pollutant, multimedia standard that is ecologically relevant and reflects the combined impacts of these two pollutants as they deposit to sensitive aquatic ecosystems.

The final *Policy Assessment* clearly sets out the basis for the recommended ranges for each of the four elements (indicator, averaging time, level and form) of a potential NAAQS that uses ambient air indicators to address the combined effects of oxides of nitrogen and oxides of sulfur on aquatic ecosystems, primarily streams and lakes. As requested in our previous letters, the *Policy Assessment* also describes the implications of choosing specific combinations of elements and provides numerous maps and tabular estimates of the spatial extent and degree of severity of NAAQS exceedances expected to result from possible combinations of the elements of the standard.

We believe this final PA is appropriate for use in determining a secondary standard to help protect aquatic ecosystems from acidifying deposition of oxides of sulfur and nitrogen. EPA staff has done a commendable job in developing the innovative Aquatic Acidification Index (AAI), which provides a framework for a national standard based on ambient concentrations that also takes into account regional differences in sensitivities of ecosystems across the country to effects of acidifying deposition.

CASAC supports the potential choices/ranges presented by EPA staff on the indicators, form, averaging time, and level that should be considered for a revised secondary NO<sub>x</sub>-SO<sub>x</sub> NAAQS. While each aspect of the standard is discussed separately in our attached comments, ultimately the final choice of the indicators, averaging time, form and level should be considered in an integrated fashion, accounting for the other considerations and uncertainties also discussed.

As EPA moves forward in this regulatory process, we recommend some attention be given to our residual concern that the available data may reflect the more sensitive water bodies and thus, the selection of percentiles of waterbodies to be protected could be conservatively biased. Further, we found some improvements could be made to the uncertainty analysis. Both of these concerns are discussed in our attached consensus comments.

In summary, while we have identified areas for improvement in future work and topics that may need to be clarified in the current review, overall CASAC finds that the *Policy Assessment* provides relevant scientific information needed for consideration in revising the secondary NAAQS for oxides of nitrogen and sulfur. We trust that our comments are useful to you and your agency in developing a proposal for a multipollutant NAAQS.

Sincerely,

*/signed/*

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

*/signed/*

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

Enclosures

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**Enclosure A: Rosters**  
**U.S. Environmental Protection Agency**  
**Clean Air Scientific Advisory Committee**  
**Oxides of Nitrogen (NO<sub>x</sub>) and Sulfur Oxides (SO<sub>x</sub>) Secondary Review Panel**

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## **Enclosure B: CASAC Consensus Comments**

Below are CASAC's detailed comments on each aspect of a potential NAAQS standard.

### **Indicators**

The use of total reactive oxidized nitrogen ( $\text{NO}_y$ ) and sulfur oxides ( $\text{SO}_x$ ) as atmospheric indicators of oxidized nitrogen (N) and sulfur (S) atmospheric concentrations is well justified. The use in the Aquatic Acidification Index (AAI) of  $\text{NO}_y$  and  $\text{SO}_x$  as atmospheric indicators of N and S concentrations is useful and corresponds with other efforts by EPA. As we have stated previously, CASAC also agrees that acid neutralizing capacity (ANC) is the most appropriate ecological indicator of aquatic ecosystem response and resiliency to acidification. It should be noted, however, that for some ecosystems, ANC does not fully characterize acidification attributes of systems and additional parameters such as an indicator of acidity or alkalinity (pH), dissolved organic carbon (DOC) and monomeric aluminum are important. While CASAC supports the use of the proposed  $\text{SO}_x$  and  $\text{NO}_y$  indicators, in combination with associated deposition transfer ratios from the Community Multiscale Air Quality (CMAQ) model, we encourage the Agency to exercise flexibility in considering alternative combinations of measured S and N chemical species and transference ratios that may be capable of producing comparable deposition estimates. The Air Monitoring and Methods Subcommittee report, (EPA-CASAC-11-006), provides comments on the methods and measurements for implementing a new secondary NAAQS for oxides of N and S. Final decisions on the N and S indicators will need to take their recommendations into consideration.

### **Form**

EPA has developed the AAI, an innovative "form" of the NAAQS itself that incorporates the multi-pollutant, multi-media, environmentally modified, geographically variable nature of  $\text{SO}_x/\text{NO}_y$  deposition-related aquatic acidification effects. With the caveats noted below, CASAC believes that this form of the NAAQS as described in the final Policy Assessment is consistent with and directly reflective of current scientific understanding of effects of acidifying deposition on aquatic ecosystems.

The form described consists of two general parts - a chemical component, and a spatial component. The chemical component, expressed in terms of the AAI equation, links an environmental indicator (a protective level of ANC in surface waters) to measurable concentrations of  $\text{SO}_x$  and  $\text{NO}_y$  in the ambient air. For any given spatial area and specified level of the standard, compliance can then be directly determined from new and continuing measurements of  $\text{SO}_x$  and  $\text{NO}_y$ . Given the importance of CMAQ and ecological model calculations in the equation, there is a continuing need to evaluate model performance in estimating the deposition terms and ecological components of the AAI to account for model uncertainties, and to make the model-dependent factors in the NAAQS increasingly transparent. Model performance issues are addressed further below.

The role of DOC and its effects on ANC, including differences in measured and calculated ANC values would benefit from further refinement and clarification. Some consideration should be

given to how the supply of DOC affects overall sensitivity of aquatic ecosystems to total acidifying deposition, and the use of ANC as an indicator of surface water acidification. In future considerations of the standard, accounting for aluminum availability and sulfur retention and/or mobilization in soils also should be considered.

CASAC agrees that the spatial components of the form in the Policy Assessment are reasonable and that use of Omernick's ecoregions (Level III) is appropriate for a secondary NAAQS intended to protect the aquatic environment from acidification – especially where there are geographical variations in the inherent sensitivity of ecosystems to pollutant effects. When considered in isolation, it is difficult to evaluate the logic or implications of selecting percentiles (70<sup>th</sup> to 90<sup>th</sup>) of the distribution of estimated critical loads for lakes in sensitive ecoregions to determine an acceptable amount of deposition for a given ecoregion. However, when these percentile ranges are combined with alternative levels within the staff-recommended ANC range of 20 to 75 microequivalents per liter ( $\mu\text{eq/L}$ ), the results using the AAI point to the ecoregions across the country that would be expected to require additional protection from acidifying deposition. Reasonable choices were made in developing the form. The number of acid sensitive regions not likely to meet the standard will be affected both by choice of ANC level and the percentile of the distribution of critical loads for lakes to meet alternative ANC levels in each region. These combined recommendations provide the Administrator with a broad but reasonable range of minimally to substantially protective options for the standard.

The division of ecoregions into “sensitive” and “non-sensitive” subsets, with a more protective percentile applied to the sensitive areas, also seems reasonable. However, it is not entirely clear how the various sensitivity criteria will actually be defined and applied. The most relevant questions to answer are whether these areas currently suffer adverse effects to public welfare from anthropogenic-caused deposition and whether there is good scientific reason to expect improvement in the condition of these areas if anthropogenic deposition were decreased. There is likely to be some geographical variability in the capacity of soils to take up (and release) atmospheric sulfur and nitrogen that could be considered as part of the sensitivity criteria. Very low calculated critical loads that result from natural acidity are not an accurate reflection of the potential for adverse effects to public welfare from anthropogenic-caused deposition. Natural poorly buffered coastal plain areas are initially identified as sensitive areas based on low ANC, but are later identified as anomalous. These areas are found in CMAQ model runs that use a single emission reduction scenario to be “non-responsive” to decreases in SO<sub>x</sub> and NO<sub>x</sub> deposition. While consideration of “responsiveness” is one of several screening criteria that might logically be applied, a more thorough assessment of responsiveness based on multiple emission/deposition-control scenarios and other factors would be helpful. The proposed rule should clearly specify how these or other screening criteria will be applied in assigning ecoregions to “sensitive” and “non-sensitive” categories.

Critical load calculations can be made either using the Model of Acidification of Groundwater in Catchments (MAGIC) or the F-factor approach. The critical loads calculated using the F-factor approach are consistently lower than those calculated using the MAGIC model. Either approach may result in an underestimate of the critical load component in the AAI equation in some regions. Calculations of critical loads for individual water bodies within each region can be accomplished using available data. The percentile of water bodies to select to meet the target

ANC value is based on this distribution. The representativeness of the available data for an ecoregion is therefore important. The available data are more likely to reflect the more sensitive water bodies and are therefore not representative of the entire population of water bodies in a given ecoregion. Potential biases that would result from using the F-factor approach and the currently available water chemistry data should be identified and addressed.

The potential biases in the critical load calculations and in the regional representativeness of available surface water chemistry data suggest that a given percentile of the distribution of estimated critical loads for surface waters may be protective of a higher percentage of surface waters in some regions. As indicated above, it is important that the performance of both atmospheric and ecological models on which the potential NAAQS is based be continually evaluated and refined. Periodically conducting direct measurements of ANC and other modeled chemical parameters in surface waters would also be a valuable way to assure that the combination of measured and modeled components of the NAAQS are performing as expected.

### **Averaging time**

Considering the cumulative nature of the long-term adverse ecological effects and the year-to-year variability of atmospheric conditions (mainly in the amount of precipitation), CASAC concurs with EPA that an averaging time of three to five years for the AAI parameters is appropriate. A longer averaging time would mask possible trends of AAI, while a shorter averaging time would make the AAI being more influenced by the conditions of the particular years selected.

### **Level**

CASAC agrees with EPA staff's recommendation that the "level" of the alternative AAI standards should be within the range of 20 and 75  $\mu\text{eq/L}$ . We also recognize that both the "level" and the form of any AAI standard are so closely linked in their effectiveness that these two elements should be considered together.

### **Other Considerations**

Adverse effects: Providing protection from adverse effects to public welfare is the motivation for this standard setting process. There is clear evidence that such adverse effects are occurring in sensitive ecosystems in the United States as a result of anthropogenic acidic deposition. There are uncertainties about the magnitude of the economic value of the losses in ecosystem services, but CASAC believes that adverse effects to public welfare exist when fish species and other aquatic life that are native to an ecosystem are unable to survive.

Process for updating the AAI: EPA should provide values for the four elements of the AAI for each ecoregion as part of the proposed rule. These values may differ across the regions, but nationally consistent methods for their determination should be used. The suggestion was made that in some cases individual states might want to adjust some of these values with their own measurement data, but CASAC is concerned that this would be problematic unless the state's measurement data are rigorously quality assured. However, it is extremely important to

recognize that during each review cycle for the standard, new ecological and atmospheric data and modeling approaches should be considered to update and improve the values for the elements of the AAI, and that the national monitoring programs that supply data for these efforts are continued and, in some cases, expanded (e.g., National Acid Deposition Program, Clean Air Status and Trends Network, Temporally Integrated Monitoring of Ecosystems, Long Term Monitoring). This includes the estimates of ammonia and ammonium ion deposition, data and models used in the critical load calculations, data and models used to determine the deposition transference ratios, the retention rates for N and S, and weathering and runoff data.

Chemically Reduced Nitrogen: Another issue raised in the context of the Aquatic Acidification Index (AAI) is the matter of chemically reduced forms of nitrogen ( $\text{NH}_x$  = sum of  $\text{NH}_3$  and  $\text{NH}_4^+$ ). CASAC fully understands that these chemically reduced forms of reactive nitrogen have not been identified as criteria pollutants. Nevertheless, aquatic ecosystems are well known to respond to these reduced forms of nitrogen as well as to oxidized nitrogen and to create acidity in the process, just as the deposition of nitric acid does. While policy and political considerations require different treatment of chemically reduced and chemically oxidized forms of nitrogen, the ecosystems upon which they fall do not so distinguish. Thus, from a scientific perspective, it is very important to bear in mind that any controls on emissions of chemically oxidized forms, which originate primarily from fossil fuel combustion sources, will not address inputs of chemically reduced forms of reactive nitrogen, which originate primarily from animal operations and agricultural sources.

## **Uncertainties**

It is difficult to judge the adequacy of the uncertainty analysis performed by EPA because of lack of details on data inputs and the methodology used, and lack of clarity in its presentation. Because of the limitations of the uncertainty analysis, it is difficult to see how this could be used to inform the choices for the standard.

In its analysis, EPA ignored some aspects of the variability in the parameters within an ecoregion. This omission could be critical in cases where this variability also affects the degree of confidence in a given value of a parameter. Instead of using the mean ecoregion values and the unexplained “bootstrapping” method to assign uncertainty, EPA could have used the variability of the distribution of the values in an ecoregion. If the distribution of values in an ecoregion is reasonably homogeneous, the effect of variability would be minimal. Under these conditions, the degree of confidence in the response surfaces for  $\text{SO}_x$  and  $\text{NO}_y$  is influenced solely by the uncertainty in the measurements or estimations of the AAI parameters. EPA’s uncertainty analysis does not include the impact of variability in judging the degree of confidence in the response surfaces by assuming average conditions and defining fixed levels of uncertainty for the average value of the parameters. In the future, EPA could perform uncertainty analysis at the water body level to account for variability of water bodies within an ecoregion to complement the analysis done for the ecoregion averages.

The parameters of the AAI equation are derived using air quality and aquatic models. Given that these models are being used in the standard setting process, a more rigorous model evaluation should have been conducted to provide more confidence in the use of the models. For example,

CMAQ model evaluation statistics averaged over the continental U.S. are shown for modeled quantities which limit the understanding of how well it simulates deposition in acid-sensitive spatial areas of interest. The MAGIC model was evaluated by EPA for 104 water bodies, but results are shown only for four water bodies. It is important to know model results for the remaining 100 water bodies.

### **Recommendations for Future Research and Analysis**

The innovative nature of this form of a combined NO<sub>x</sub>-SO<sub>x</sub> secondary NAAQS has prompted CASAC to identify a number of areas that should be the focus of further research. These areas are detailed here and have been documented in past transmittals, including the attached panelists' comments. In many cases, EPA staff has been able to address the issues identified. Particular areas that would benefit from further study or consideration in potential revisions or modifications to the form of the NAAQS include sulfur retention and mobilization in the soils, aluminum availability, soil versus water acidification, and ecosystem recovery times. The important role that chemically reduced and organic forms of total reactive nitrogen have, not only in ecosystem acidification, but also in excess nutrient loading suggests that EPA identify ways to limit future emissions of reduced nitrogen (e.g., ammonia). Although the current focus on an ecological indicator of acidification effects on aquatic ecosystems is appropriate, some further linkage with associated effects on terrestrial ecosystems would be helpful in future work.

The use of the chemiluminescence technique for measuring, NO<sub>y</sub>, has considerable merit. However, monitoring of individual ambient N chemical species that is more consistent with the current understanding of atmospheric deposition processes is also recommended for evaluation of dry deposition values generated by CMAQ. Nitric acid (HNO<sub>3</sub>), nitrogen dioxide (NO<sub>2</sub>) and ammonia gas (NH<sub>3</sub>) are routinely and reliably monitored with passive samplers. Since these species, in part, account for the majority of the dry deposited N, a commitment to their monitoring is encouraged. These measurements are especially important for reliable estimates of NH<sub>x</sub> deposition (factor F2 in the equation for calculation of AAI). Such empirical data would also help in calculation of the transference ratios from CMAQ model simulations. Ambient air monitoring with passive samplers in conjunction with the Clean Air Status and Trends Network (CASTNET) measurements, would greatly improve robustness of the available air chemistry data that would be essential for evaluation of the CMAQ outputs. The *in situ* measurements of total N & S deposition at a subset of monitoring sites are also recommended for evaluating CMAQ. An accompanying letter based on the work of the Air Monitoring and Methods Subcommittee will provide additional comments on these measurements.

**Individual Comments from the Panel**  
**Policy Assessment for the Review of the Secondary National Ambient Air Quality**  
**Standards for Oxides of Nitrogen and Oxides of Sulfur (2011)**

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**Dr. Praveen Amar**

**General Comments:**

First, EPA staff is to be commended for producing an outstanding document. This version of the Policy Assessment Document (PAD) is extremely well-written and articulates beautifully many complex scientific concepts of atmospheric processes, ecological processes, and ecological effects (both chemical and biogeochemical). The PAD is written in a highly policy-relevant language in that it makes an excellent and convincing case for the need for establishing new secondary National Ambient Air Quality Standards (NAAQS) for protection of sensitive aquatic ecosystems in the United States. It also makes the case for retaining the existing secondary NAAQS for SO<sub>x</sub> and NO<sub>x</sub> to protect against the known direct adverse effects.

The proposed standard in this final PAD is expressed as a simple equation that explicitly takes into account the multi-pollutant nature of chemical species responsible for the adverse effects (NO<sub>y</sub>, NH<sub>x</sub> (only indirectly, see below), and SO<sub>x</sub>), as well as multi-media nature of ecological effects (aquatic ecosystems (in this effort) and terrestrial ecosystems (in the future), both acidification (in this effort) and nitrogen nutrient enrichment effects (future effort). As a first order, the equation for AAI (Aquatic Acidification Index):

$$AAI = F1 - F2 - F3 (NO_y) - F4 (SO_x)$$

is the “form” of the “NAAQS” itself? The recommended indicator, ANC, is ecological in nature (and, not an “in-the-air” variable). Acid neutralizing capacity (ANC) is presented as the best measure of the capacity of an aquatic ecosystem to protect against acidifying deposition and has been strongly supported by our CASAC Panel in our previous deliberations and comments. The range of averaging time of three to five consecutive years for calculating average annual AAI values is consistent with the cumulative nature of the long-term adverse ecological effects, and this long-time average should be able to account for the generally large interannual variability of wet precipitation (rain, snow). It is also in agreement with our previous CASAC deliberations and comments.

In the future “real-world” implementation of the standard, the calculated values of AAI for a given spatial area (Omernick’s Ecoregion (Level III)) will be compared to the level of the AAI standard, yet to be chosen. The numerical value of “AAI standard” is directly related to the ecological indicator, ANC, and once the exact level of ANC (within the recommended range of 20 to 75 micro equivalents per liter) is chosen, it determines the numerical values of the AAI standard. It is expected that EPA will provide/codify ecoregion-specific “look-up” tables to state/ local/regional/tribal agencies and other interested parties with values for all of the four factors (F1, F2, F3, F4), included as a part of the proposed Rule.

To make a determination as to whether the ‘standard’ is being met, one would simply need the measured air concentrations of NO<sub>y</sub> and SO<sub>x</sub> in a given ecoregion, together with the EPA-provided values of four ecoregion area-specific F1-F4 factors into the equation for AAI, and then compare resulting AAI value to the level of the standard. EPA should be asked for making

this simplified approach available to implementing state/local/regional/tribal entities *right in the Rule* as it is promulgated. Otherwise, the rather complex nature (though entirely necessary and based on some beautiful, elegant, and well thought-out science) of this NAAQS, compared to the all the previous “simple” NAAQS (both primary and secondary) will result in technical, institutional, educational, public-outreach, and “cultural” barriers that may be difficult to overcome.

The complex nature of various components (and their variability over space and time as well as uncertainty in parameters as well as models, both atmospheric and ecological) requires that the approach finally selected is “more” than the sum of its parts. The suggested AAI approach, with its chemical components and spatial regions over which these components are averaged, is indeed “more” than the sum of its individual parts and does a very good job of providing a rationale for secondary NAAQS.

It appears that the AAI framework has gone as far as it could go in addressing the reduced nitrogen (ambient NH<sub>x</sub> and its deposition). However, the framework only “takes into account” NH<sub>x</sub> to calculate what levels of NO<sub>x</sub> and SO<sub>x</sub> emission reductions might be required in the future to meet the secondary NAAQS based on AAI. Thus future control strategies and policy options most probably will not allow federal EPA (or, for that matter, states) to address and require reductions in U.S. ammonia emissions to meet the AAI standard. In this sense, it is a missed opportunity to directly and explicitly address ammonia emissions in the U.S., which unlike SO<sub>x</sub> and NO<sub>x</sub> emissions are currently *increasing* because of increased food production and increased activity in CAFO (confined animal feeding operations) sources.

On Page 7-38, the PAD suggests that because “NH<sub>x</sub> deposition exhibits greater spatial variability, as well as overall uncertainty than other terms,” an alternative approach might involve the use of more localized and/or contemporaneous modeling. Other approaches might involve the use of monitored NH<sub>x</sub> data. The other parts of the document suggest that states may choose alternative approaches for addressing NH<sub>x</sub> emissions and ambient levels. For the sake of consistency in any future implementation of the AAI standard, I recommend that a uniform national approach must be developed instead of the state-by-state approach.

### **Consideration Associated with Alternative Standards:**

It is not clear how the stated intent of the AAI standard to average AAI over three to five consecutive years would be actually met because there is “the lack of CMAQ modeling for multiple consecutive years.” (page 7-54).

The PAD addresses the “responsiveness” of certain ecoregions (page 7-62, also see page 7-64, where “responsiveness to deposition change” is noted as one of the four parameters in delineating an ecoregion as relatively non-acid sensitive) to future reductions in SO<sub>x</sub> and NO<sub>x</sub> emissions. It makes a statement (page 7-62) that “expected emission changes over the next two decades should be *far greater* (my emphasis) than the 42% and 48% SO<sub>x</sub> and NO<sub>x</sub> reductions used in this analysis.” I do not think, based on the “rules on the book,” and general historic trends, and considering emissions from all sources (and, not just power plants), this statement is

true. The expected reductions might even be smaller. Recognizing that “responsiveness”/emission sensitivity is used as one of the parameters (as noted above) to estimate certain ecoregions “likely not meeting alternative standards,” it may be important to evaluate the statement made next “with a consequent further reductions in ecoregions that would likely not meet alternative standards.”

Finally, Section 7.6 on system uncertainties is an excellent summary of various new analyses done to undertake a rather rigorous and complete analyses of uncertainty, variability, and sensitivity in a common-sense manner.

## Dr. Andrzej Bytnerowicz

### Comments

Generally this version of the Policy Assessment is a greatly improved document compared with its previous versions. I would like to complement the EPA staff for their efforts and persistence in developing the Policy Assessment that very well describes and justifies the rationale for developing a secondary standard for  $\text{NO}_x$  and  $\text{SO}_x$ . I support the proposed approach for implementing the ecologically-based new secondary standard for  $\text{NO}_x$  and  $\text{SO}_x$  that is based on the best available science and creative use of models.

#### Chapter 7. Consideration of alternative standards for aquatic acidification

I have been specifically asked to comment on indicators described in this chapter. I have also made comment on other related issues discussed in this chapter. These comments mainly focus on the air chemistry part of the proposed methodology. I also make some suggestions for improving the existing national monitoring networks and methodologies.

Chapter 7 is well organized and is written clearly and concisely. The introductory section provides a convincing reasoning why the new standard is needed and good overview of the proposed methodology. It also provides a justification why the present approach for multiple pollutants has been developed; why only the aquatic ecosystems have been selected for the secondary standard; considerations for using specific values of AAI for various ecological zones across the country; and considerations for the averaging time of the standard.

I agree that at present using the total reactive oxidized nitrogen,  $\text{NO}_y$ , measured with the chemiluminescence technique is justified. However, as the authors state on page 7-5, using individual ambient N species would be more consistent with the current understanding of atmospheric deposition science and has a better potential for more rigorous evaluation of dry deposition values. Although  $\text{HNO}_3$  is the strongest driver of N dry deposition, it is not a good indicator of dry deposition by itself (as the EPA has shown), but together with  $\text{NO}_2$  and  $\text{NH}_3$  it accounts for the majority of the dry deposited N. These chemical species can be routinely and reliably monitored with passive samplers. If these species are measured in conjunction with the CASTNET filter pack sampler, much more reliable data of  $\text{HNO}_3$ ,  $\text{NH}_3$  as well as particulate  $\text{NO}_3$  and  $\text{NH}_4$  concentrations from the network could be obtained. I will provide more detailed explanation of this proposed approach in the February 16<sup>th</sup> discussion on the monitoring methodologies that could support the new secondary standard for  $\text{NO}_x$  and  $\text{SO}_x$ . However, there are some pollutants, such as PAN as well as other organic N pollutants that are not easy to monitor on the regular basis. New approaches to monitoring of these pollutants should be comprehensively discussed. Considering all those technical problems and data gaps, the proposed use of CMAQ to provide the modeled values of N and S deposition is currently justified. However, the well-coordinated national efforts are needed to assure that the modeled values are verified against the experimental results. In this regard the passive samplers as well as the remote sensing techniques seem to be the most promising methodologies in the near future. Potential utilization of these techniques is especially important for  $\text{NH}_x$  – even if the agricultural sources are better characterized in the CMAQ predictions, emissions from multiple mobile sources (combustion engines) and their spatial trends are very difficult to model. Therefore, reliable monitoring of the ambient  $\text{NH}_3$  and  $\text{NH}_4$  at a national scale is urgently needed.

There is also a clear need for monitoring coarse particulate matter (PM) which is an important source of the atmospheric N and S into the aquatic and terrestrial ecosystems. Consequently, developing of monitoring techniques for coarse PM is recommended.

In regard to choosing ecological indicators, a selection of the ANC is logical, well described and justified. The use of ANC has already been discussed at length between the EPA staff and the CASAC. This indicator is based on solid science and is well related to the combined deposition of  $\text{NO}_y$ ,  $\text{SO}_2$  &  $\text{SO}_4$  and  $\text{NH}_x$ .

The transference ratios have to be verified since they are just products of the model. In this regard, the *in situ* measurements of total deposition are needed. The ion exchange resin collectors, which have been used both in the East and the West, could be considered as a reference method for selected watersheds. These measurements should be matched with reliable measurements of the individual components of the  $\text{NO}_y$ .

In regard to the spatial aggregation, it is clearly stated that variation in the ecosystem sensitivity to N & S deposition has to be included in the development of a national standard. Selection of the Omernik's classification with 84 defined regions seems logical. Remark – the text on pages 7-32 and 7-33 as well as the caption for Figure 7.5 has to be checked since the numbers for the ecoregions and subdivisions are not consistent. Similarly – the last paragraph on page 7-34 is confusing and does not provide any explanation why the selected percentages of water bodies and ANC values were used to screen out regions with the overabundance of the high ANC values.

The summary of system uncertainties (Section 7.6) and Table 7-6-1 with results of the uncertainty analysis are informative and useful. What I have found most interesting is that the highest uncertainties are caused by the transference ratios and the Neco. This fact clearly indicates that more information from the *in situ* atmospheric deposition measurement and the ecological effects evaluations are needed. It is especially important considering that the changing climate will provide additional uncertainties in regard to these parameters in future years. Therefore, sufficient efforts have to be undertaken to assure that the proposed methodologies are adequately tested and compared with the actual field measurements.

Table 7-6-1. “Summary of qualitative uncertainty analysis of key components of AAI”.

(a) In regard to the “atmospheric concentrations to deposition” source of uncertainty, I have problems believing that the “knowledge –base uncertainty” is “low”. In the comments of the same table there is a statement that greater uncertainties reside in the information driving these calculations, such as the  $\text{NH}_3$  emissions. Therefore verification against the ground-level measurements (see my comments above) is needed. Without such comparisons I would be hesitant to agree that the uncertainty for this factor is “low”.

(b) In regard to “Dep  $\text{NO}_y$ ” & “Dep  $\text{NH}_x$ ” and the related comments in the table: again, testing of the models against ambient  $\text{NO}_y$  and  $\text{NH}_x$  experimental data is essential. I believe that the CASTNET results aided by the passive sampler data, the Ncore network and possibly in the remotely sensed data can be very useful in this regard.

Appendix D containing Figures D-1 through D-9 and Tables D-3 & D-6. Information contained in those figures and tables is quite educational and can greatly help the Administrator in deciding what level of protection should be selected. Clearly, setting a standard at 75 ueq/L and 90<sup>th</sup> percentile provides the best protection. Comparing the present scenarios with those with the reduced emissions shows benefits of the stricter  $\text{NO}_x$  &  $\text{SO}_x$  air pollution regulations.

## Ms. Lauraine Chestnut

This draft final of the Policy Assessment document is much improved over the last draft CASAC reviewed. It is responsive to comments on that draft. It presents the rationale and approach proposed for the NO<sub>x</sub>-SO<sub>x</sub> secondary NAAQS in a more coherent and comprehensive manner, making it easier to understand without getting bogged down in the many technical details.

I think there is sufficient scientific support for proceeding with the proposed secondary standards to protect aquatic resources from the effects of acidic deposition. This draft of the Policy Assessment addresses three important questions that were raised during the previous review:

1. Completion of the staff's recommendations as to appropriate ranges for the indicator, form, averaging time, and level of the standard
2. A more thorough assessment of uncertainties and their implications for the application of the AAI
3. Examples of how the AAI would be calculated and applied for each ecoregion

I have some remaining concerns and questions on some specifics in the document.

### **Evidence regarding adversity of effects**

This draft of the PA does a good job summarizing this evidence and how it applied in this case. I just have a couple minor issues. One is that uncertainty in the *magnitude* of the economic value of the effects on ecosystem services is not the same thing as uncertainty about whether the effects at current levels are adverse to public welfare. When effects are at the level where entire species of fish are unable to survive in their native waters as a result of human produced pollution, there is very little uncertainty that these effects are adverse to public welfare. This could be more strongly stated in the conclusions of Chapter 7. It is unnecessary to focus so much on the admittedly substantial uncertainty about the magnitude of the economic value of the effects on public welfare, for the purposes of setting this standard.

A minor note: I am still unclear about the differences between the estimates in Tables 4-3 and 4-4 with regard to the different ANC thresholds.

### **Water quality data and selection of percentiles for calculation of alternative critical loads for an ecoregion**

My understanding is that specific measures of water quality are needed for each water body in an ecoregion to calculate a critical load for that water body (for a given ANC target). Using all available data for water bodies in an ecoregion, a distribution of critical loads for the ecoregion is then obtained. From this distribution a critical load for a selected percentile of the population of water bodies in the ecoregion can be selected. This process presumes that the water quality data available are for a representative sample of water bodies in the region. I don't know enough about the sources of these data to assess how reasonable this presumption is. If data are collected only for water bodies suspected of being sensitive to acidification, for example, then the data are not going to represent the whole population of water bodies in an ecoregion. In that case it is hard to know what a percentile of that distribution means. An even more troubling concern is that if data available for different ecoregions are based on different types of samples of water bodies, then there is no comparability in terms of what a given percentile means across the different ecoregions.

It may be appropriate to treat these water quality data as something that can be updated or substituted with improved data as they become available (or if states want to invest in better data collection); similar to what is proposed for handling reduced N.

**Differentiation in the treatment of acid-sensitive and not acid-sensitive ecoregions**

There are two issues here. One is that the rationale for using 50<sup>th</sup> percentile critical load for ecoregions that are not sensitive to acidification (i.e., with high buffering capacity) versus 70<sup>th</sup> to 90<sup>th</sup> for sensitive ecoregions is not entirely convincing. If they are not sensitive, then wouldn't the critical load at the 70<sup>th</sup> or 90<sup>th</sup> percentile still be quite high? I'm worried about stretching things when this is supposed to be a national standard.

The second issue is the proposal that Atlantic coastal areas be treated as not sensitive because they appear to be unresponsive to large reductions in deposition levels. Is the scientific evidence convincing that these regions are naturally acidic and are therefore not adversely affected by human produced deposition? If so, then why were these regions not captured in the initial screening of naturally acidic areas?

**Time to recovery**

The PA concludes that it is not possible to say anything about how long it would take for various ecoregions to recover with reduced deposition, other than to say the lower the deposition the more rapid the recovery. Maybe this is all that can be said, but it remains a policy-relevant question. Leaving it so open-ended leaves room for the argument that it is not worth investing much today for a recovery that won't be seen until many decades in the future. Can nothing more specific be said based on evidence from the past 10-15 years with the Clean Air Act Amendments?

**Dr. Ellis Cowling**

Chairman Russell asked all members of this CASAC NO<sub>x</sub>/SO<sub>x</sub> Secondary NAAQS Review Panel to provide general comments “on any and all aspects” of the January 14 Policy Assessment and some of us to offer more specific comments on one or another topic within Chapter 7 titled “Consideration of Alternative Standards for Aquatic Acidification.” Both Dale Johnson and I have been asked to address the “Level” of the proposed alternative and integrated NO<sub>x</sub> and SO<sub>x</sub> NAAQS Secondary Standards.

Accordingly, please find below both some general comments on the January 14, 2011 Policy Assessment document and some more specific comments on the recommended “Level” of the proposed integrated NAAQS Secondary Standard for NO<sub>x</sub> and SO<sub>x</sub>.

**General Comments:**

EPA Staff have come a very long way toward their own self-declared objective of developing a scientifically sound “framework for a multi-pollutant, multimedia standard that is ecologically relevant and reflects the combined impacts of these two pollutants [total reactive nitrogen and sulfur] as they deposit to sensitive aquatic ecosystems!”

I applaud EPA staff’s significant achievements in the analysis and interpretation of current scientific understanding of atmospheric-deposition induced effects on aquatic and terrestrial ecosystems and in the development of science-based policy assessments and recommendations in the following areas of public welfare protection:

- 1) Recommending that the present separate NO<sub>2</sub> and SO<sub>2</sub> NAAQS Secondary Standards should be retained to continue to provide protection for vegetation from direct damage associated with exposures to gaseous SO<sub>2</sub> and NO<sub>2</sub>.
- 2) Recognizing that these existing standards are neither appropriate nor adequate to provide protection of fresh water aquatic ecosystems from atmospheric deposition of total reactive nitrogen and sulfur compounds that are causing significant adverse public welfare impacts in acid sensitive in various regions across the United States.
- 3) Recognizing that both total reactive nitrogen and sulfur compounds are the principal causes of atmospheric-deposition-induced acidification effects on aquatic ecosystems and that a multiple pollutant approach to air quality management is essential to provide adequate protection of these ecosystems in various acid sensitive regions of the United States.
- 4) Developing a scientifically sound framework for relating air emissions of well characterized sulfur and nitrogen pollutants, to measurable ambient air concentrations of total reactive nitrogen and sulfur compounds, to atmospheric deposition of total acidifying deposition, doing this in reasonably well-defined acid-sensitive and non-acid sensitive regions of the US, where specific types of damage to fish and other aquatic biota and various other kinds of adverse impairments of ecosystem functions are occurring, and where acid neutralizing capacity (ANC) is universally

recognized as the most reliable measure of aquatic ecosystem sensitivity to both chronic and episodic acidification processes, and where many of the factors that determine the time-frame and spatial distribution of biological response to decreases in allowable nitrogen and sulfur emissions and atmospheric deposition are reasonably well understood.

4) Development of the concept of an Aquatic Acidification Index (AAI) and a relatively simple mathematical equation by which to calculate and quantify the changes in nitrogen and sulfur pollutant emissions that will be needed to provide adequate protection for acid-sensitive aquatic ecosystems in various regions of the US.

5) Recognizing that the base of scientific understanding with regard to nitrogen- and sulfur-induced acidification effects on aquatic ecosystems is very much more substantial than the science base for understanding N and S induced nutrient enrichment effects on aquatic ecosystems and also for understanding both acidification and nutrient enrichment effects on terrestrial ecosystems.

Thus concluding that it is best (more wise in terms of public policy formulation) to begin an integrated multi-pollutant and multiple-ecological effects program by concentrating (at least for now) on development and implementation of a secondary NAAQS standard to protect aquatic ecosystems from acidification and to pursue further scientific research and assessment efforts before including nitrogen enrichment effects on aquatic ecosystems and either acidification and nitrogen enrichment effects on terrestrial ecosystems.

But also believing that the conceptual framework outlined in Item 3 (above) may provide a useful foundational framework that could later be adapted for use in dealing with both nitrogen enrichment effects on aquatic ecosystems and both acidification and nitrogen enrichment effects on terrestrial ecosystems.

6) Developing a policy assessment document that presents these many interrelated concepts and both atmospheric and ecological processes in a clear and logical fashion with very specific and clear presentation of the chemical indicators of choice, the statistical form and averaging time that would be appropriate, and the range of ANC levels that would be appropriate for consideration, promulgation and implementation to significantly decrease the presently occurring substantial adverse effects on public welfare in various regions of the United States.

### **Specific Comments:**

With regard to the Level of an Integrated NAAQS Secondary Standard I am persuaded by the ideas developed by EPA staff on pages 7-44 through 7-53 (including earlier advice that CASAC offered in its response to review of the Second External Review Draft of the Policy Assessment) that “target ANC levels in the range of 20 to 75 $\mu$ eq/L” are appropriate for consideration by EPA Administrator Jackson. As pointed out on pages 7-50 and 7-51, this range of target ANC values is consistent with those that have been implemented by other organizations including the UNECE in Europe, and the states of New York, Vermont, New Hampshire, and Tennessee in this country.

## Additional Concerns:

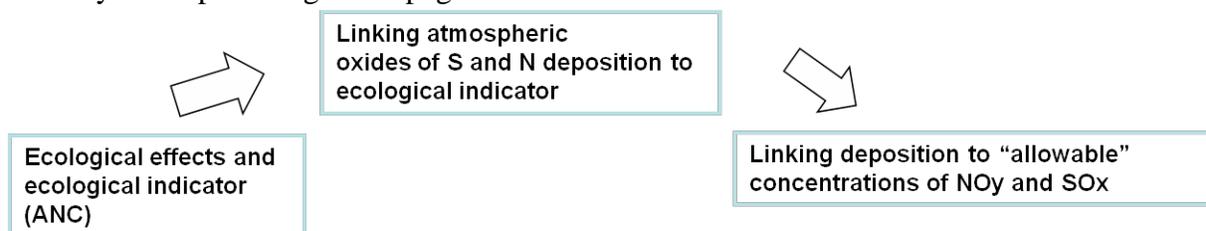
With regard to public understanding of the concepts of total acidifying deposition and total reactive nitrogen in the context of further consideration of the alternative NAAQS standards developed in this nearly final Policy Assessment document, however, I am greatly concerned that the general tenor, focus, and content of this document will continue to lead the US public as a whole to continue to believe in the **myth that it is only oxides of nitrogen and sulfur that are really injurious to public health and public welfare in this country.**

I am well aware that chemically reduced forms of reactive nitrogen are acknowledged and dealt with specifically as part of the description of total acidifying deposition that is the root cause of the acidification of aquatic ecosystems to which this whole Policy Assessment document is addressed. But the more or less constant and repeated use of use of the terms “oxides of nitrogen in the ambient air,” “nitrogen and sulfur oxides,” etc., and the comparatively infrequent use of the term “total reactive nitrogen” and “total acidifying deposition” leads to perpetuation of the myth that it is oxidized forms of nitrogen and sulfur that are the real causes of injury to public health and environment.

Consider the following statement on the first page of the Executive Summary:

“We have a high degree of confidence in the linkages between atmospheric oxides of nitrogen and sulfur, associated deposition of nitrogen and sulfur, and deposition-related aquatic acidification effects. Our objective in this Policy Assessment is to develop a framework for a multi-pollutant, multimedia standard that is ecologically relevant and reflects the combined impacts of these two pollutants as they deposit to sensitive aquatic ecosystems.”

Or also consider the repeated use of the diagram first shown on page ES-6 in the Executive Summary and repeated again on pages 7-9 and 7-26:



**Conceptual design of the form of an aquatic acidification standard for oxides of nitrogen and sulfur.**

Would it not be more appropriate (and more truthful!) for this very significant conceptual diagram to be reworded by: 1) leaving out the misleading words in the middle box of his diagram that refer only to “oxides of S and N,” 2) potentially changing the last line of the right-hand box to read “concentrations of NO<sub>y</sub>, SO<sub>x</sub>, and NH<sub>x</sub>,” and 3) changing the caption to read:

**Conceptual design of the form of an aquatic acidification standard that includes all ecosystem-acidifying forms of nitrogen and sulfur.**

I raise this general question about the potential of this otherwise very well-conceived and well-written Policy Assessment document to serve an important **public educational purpose as well as a Policy Assessment purpose** in very large part because of the following statements that are made in various chapters of this Policy Assessment document:

Page 7-78 --“In considering an indicator for oxides of nitrogen, we recognize that aquatic acidification results from and is best understood in terms of the deposition of total nitrogen, in both oxidized and reduced forms. Since the pollutant that is the focus of this review is oxides of nitrogen, not reduced forms of nitrogen, we conclude that it is appropriate to consider reduced forms of nitrogen separately, as a factor in the form of the standard, rather than as part of the indicator of the standard.”

Page 6-5 -- “ ... it is important that the structure of the [proposed] standards address the role of reduced nitrogen in determining the ecological effects resulting from deposition of atmospheric oxides of nitrogen and sulfur.”

Page 2-15 – “The key pollutants for this assessment are total oxidized nitrogen (NO<sub>y</sub>), total reduced nitrogen (NH<sub>x</sub>), and total oxidized sulfur compounds (SO<sub>x</sub>) in ambient air.”

Page 2-15 – “Total reduced nitrogen (NH<sub>x</sub>) includes ammonia, NH<sub>3</sub>, plus ammonium, NH<sub>4</sub> (EPA, 2008) is introduced because NH<sub>x</sub> contributes to ... acidifying deposition, effectively behaving similarly to NO<sub>y</sub>.”

Page 2-15 – “Reduced nitrogen plus oxidized nitrogen is referred to as total reactive nitrogen.” –.

Page 2-38 – “The contribution of reduced nitrogen to total nitrogen deposition (Figure 2-24) illustrates the strong influence of agricultural based ammonia emissions, particularly in upper Midwest and eastern North Carolina.”

Page 3-2 – “The chemical forms of nitrogen that may contribute to acidifying deposition include both oxidized and reduced chemical species.”

Page 3-19 – “Both oxides of nitrogen and reduced forms of nitrogen (NH<sub>x</sub>) contribute to nitrogen deposition. For the most part, nitrogen effects on ecosystems do not depend on whether the nitrogen is in oxidized or reduced form.”

Page 6-5 – “There are also important interactions between oxides of nitrogen and sulfur and reduced forms of nitrogen, which also contribute to acidification and nutrient enrichment.”

Page 6-10 – “The evidence is sufficient to infer a causal relationship between N deposition, to which oxidized and reduced nitrogen contribute. .

Page 7-9 – “The [AAI] index also accounts for the contribution of reduced nitrogen to acidification.”

Pages 7-20 - 7-21 – “... we use the terms S and N in the *CLANClim* (*N + S*) term to broadly represent all species of sulfur or nitrogen that can contribute acidifying deposition. This follows conventions used in the scientific literature that addresses critical loads, and it reflects all possible acidifying contributions from any sulfur or nitrogen species. For all practical purposes, S reflects SO<sub>x</sub> as described in section 7.1, the sum of sulfur dioxide gas and particulate sulfate. However, N includes both oxidized forms, consistent with the ambient indicator, NO<sub>y</sub>, in addition to reduced nitrogen species, ammonia and ammonium ion, referred to as NH<sub>x</sub>. NH<sub>x</sub> is included in the critical load formulation

because it contributes to potentially acidifying nitrogen deposition. Consequently, from a mass balance or modeling perspective, the form of the standard must account for NH<sub>x</sub>. However, N includes both oxidized forms, consistent with the ambient indicator, NO<sub>y</sub>, in addition to reduced nitrogen species, ammonia and ammonium ion, referred to as NH<sub>x</sub>. NH<sub>x</sub> is included in the critical load formulation because it contributes to [total] acidifying nitrogen deposition. Consequently, from a mass balance or modeling perspective, the form of the standard must account for NH<sub>x</sub>.”

Page 7-28 – “Equation 7-11 is the basic expression of the standard which translates the simple conceptual diagram into an explicit expression that relates ANC as a function of the ambient air indicators, NO<sub>y</sub> and SO<sub>x</sub>. Based on equation 7-11, we define an aquatic acidification index (AAI) that is more simply stated in terms that emphasize the ambient air indicators:

$$AAI = F1 - F2 - F3[NO_y] - F4[SO_x] \quad (7-12)$$

where the AAI represents the long term (or steady state) ANC level associated with ambient air concentrations of NO<sub>y</sub> and SO<sub>x</sub>. The AAI is the potential the atmosphere affords in influencing ecosystem ANC. The factors F1 through F4 convey three attributes: a relative measure of the ecosystem’s ability to neutralize acids (F1), the acidifying potential of reduced nitrogen deposition (F2), and the deposition-to-concentration translators for NO<sub>y</sub> (F3) and SO<sub>x</sub> (F4).

Specifically:

$$F1 = AN_{Clim} + CL_r/Q_r;$$

$$F2 = NH_x/Q_r = NH_x \text{ deposition divided by } Q_r;$$

$$F3 = T_{NO_y}/Q_r; T_{NO_y} \text{ is the transference ratio that converts deposition of } NO_y \text{ to ambient air concentrations of } NO_y; \text{ and}$$

$$F4 = T_{SO_x}/Q_r; T_{SO_x} \text{ is the transference ratio that converts deposition of } SO_x \text{ to ambient air concentrations of } SO_x. .$$

F2 reflects the deposition of reduced nitrogen. Consideration should be given to specifying the value of F2 for each region based on the averaged modeled value across the region, using national CMAQ modeling that has been conducted by EPA. Consideration could also be given to alternative approaches to specifying this value, such as allowance for the use of air quality modeling conducted by States using more refined model inputs.

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PS. After making this fairly long list of places in this Policy Assessment document that did refer to the importance of chemically reduced forms of nitrogen, I couldn’t help but wonder if this fairly long list didn’t of itself argue against the very point I am trying to make -- that the general tenor, focus, and content of this document gives undue and inappropriate emphasis to oxides of nitrogen and sulfur rather than to both chemically oxidized and chemically reduced forms of nitrogen and sulfur.

Thus, I stand by my original assertion of a lack of balance in the document as a whole!

**Dr. Charles Driscoll**

I want to commend the staff of the U.S. Environmental Protection Agency (EPA) for their work on “Policy Assessment for the Review of the Secondary National Ambient Air Quality Standards for Oxides of Nitrogen and Oxides of Sulfur” (PA). The EPA staff has been very responsive to the comments and concerns of CASAC in the previous review of the PA. This draft of the PA is much clearer and transparent than the previous draft. EPA staff has clearly the indicator, form, averaging time and level that might be used for the proposed secondary NAAQS for oxides of nitrogen and sulfur.

As indicated in our previous review, EPA staff has proposed an innovative approach that utilizes the aquatic acidification index (AAI). The AAI considers atmospheric concentrations of NO<sub>x</sub> and SO<sub>x</sub>, the effects of reduced nitrogen deposition and the ability of watersheds to neutralize inputs of acidic deposition through the supply of basic cations and the retention of nitrogen.

The EPA staff conducted analysis to constrain values of the terms and conditions to apply the AAI. They showed for 19 acid-sensitive ecoregions, the ecoregions that would not meet the proposed secondary standard given alternative values of target ANC and percentile of waters to be protected. This is the type of analysis that CASAC requested in its previous review of the PA. This analysis gives CASAC and other reviewers of the PA an idea of the implications of a potential secondary standard and the extent of resources in ecoregions that are at risk with respect to acidification of freshwaters from acidic deposition.

These results of the EPA analyses make good sense given levels of atmospheric sulfur and nitrogen deposition and our understanding of acid-sensitive regions. In particular, considerable research has shown acidification effects on the Appalachian mountain areas. Less attention has been given to the coastal plains. However, these are very sensitive systems. They generally have high concentrations of naturally occurring organic acids. I believe that algorithms are available that could be applied in the AAI framework that could allow consideration of the effects of organic acids on the acid-base status of waters.

The EPA also provides some discussion of uncertainty associated with the AAI calculations. They are to be commended for doing this given the limited time available to revise the PA. This analysis provides a solid basis to conduct further uncertainty and sensitivity analysis.

In summary, although I have a few suggestions and comments, I have no difficulty supporting the PA.

**Specific comments:**

- |                       |   |
|-----------------------|---|
| ES-3, paragraph 2     | Brook trout is not a very sensitive fish species. |
| Figures throughout ES | Figure titles should be provided for figures.     |
| ES-4, line 1          | ...growing in the <u>eastern</u> United States.   |

ES-4, second paragraph	It might also be useful to refer to the results of Thomas et al. (2010) who show that there is a differential response of tree growth (some having a positive growth response; others a negative response) to increasing nitrogen deposition.
ES-6, paragraph 2, last line in paragraph	...effects in the future, including <u>effects of nutrient enrichment</u> .
7-13, eqn 7-1, Note Ca <sup>2+</sup>	This equation should also include naturally occurring organic anions.
7-18, 1 <sup>st</sup> paragraph, line 2	...dominated by precipitation <u>and evapotranspiration</u> ...
7-18, 3 <sup>rd</sup> paragraph, line 11	Do you mean less parameterization?
7-26, 1 <sup>st</sup> paragraph, 2 <sup>nd</sup> to last line	1980s
7-27	In a previous meeting, CASAC expressed concern over the assumption that no provision was given for watershed sulfur retention. I believe that a sulfur retention term, similar to Neco could be developed and applied in this framework.
7-30, 1 <sup>st</sup> paragraph	This is an important concept. It is too bad it is buried in the text. I urge EPA to pull this out and include it in the ES as well as the conclusions so the reader understands this point.
7-46, 1 <sup>st</sup> paragraph	Wow what a mess. This paragraph is horrible. Either re-write or indicate that surface waters that have been extremely impacted by acidic deposition would only have minimum ANC value of maybe -10 µeq/L. Within the ANC range of -10 to 100 µeq/L, there is a correspondence of increases in pH with increases in ANC.
	Reading this paragraph does not provide great confidence in the writer's understanding of acid-base chemistry.
7-46, 2 <sup>nd</sup> paragraph	Again, brook trout is not a sensitive species.
7-47, 2 <sup>nd</sup> paragraph	...below 50 µeq/L <u>the sensitivity of surface waters to acid inputs increases</u> and negative...

7-47, 2<sup>nd</sup> paragraph, line 12

50 µeq/L

7-52, 2<sup>nd</sup> and 3<sup>rd</sup> paragraphs

provide references for cost estimates.

7-56

It would be helpful to provide a map or a table with a better description of the sensitive ecoregions.

7-61, line 1

Do you mean Appalachian mountain areas instead of southern Appalachian mountain?

**References:**

(Thomas et al., 2010)

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

## **Dr. H. Christopher Frey**

**Chapter 7:** The chapter is generally well organized and well written and more clearly and completely conveys the indicator, form, level, and averaging time than the previous draft. The chapter also does a good job of identifying the spatial extent of the ecosystem regions that will be used.

The term “non-acid sensitive” is unclear. It implies sensitivity to a non-acid. The intended meaning appears to be ‘insensitive to acid deposition’ and thus would more accurately be ‘acid deposition insensitive’ or ‘acid insensitive.’

p. 7-37. 90<sup>th</sup> percentile usually implies that 90 percent of the population has a value less than or equal to the specified value. However, here it appears that the complementary cumulative frequency is being used. This could be made clear.

p. 7-38. The monitor coverage in each eco region is not discussed. Are there regions that have more than one monitor? If so, should they be averaged?

p. 7-48: the text here claims that CASAC’s responses on the 2<sup>nd</sup> PA “provide support for focusing on target ANC levels in the range of 20 to 75 µeq/L.” However, this statement ignores that in the letter to the Administrator, CASAC clearly stated “Acid neutralizing capacity targets in the range of 20 to 100 µeq/L appear appropriate to consider at this time.” Thus, it is unclear as to how EPA reached a conclusion based on CASAC input that the upper limit should be only 75 µeq/L rather than 100 µeq/L. In fact, even the paragraph from CASAC’s response to the 2<sup>nd</sup> PA Charge Question 11, which EPA only partially quoted in the 3<sup>rd</sup> draft PA, goes on to state: “As a result, target ANC values of 20 to 100 µeq/L are in the range of appropriate values, while recognizing that there is additional protection at 50 to 100 µeq/L”

p 7-49. The quoted text from the CASAC response to Charge Question 22.C.ii importantly refers to “long-term” ANC levels. A long-term ANC level of 75 µeq/L does not necessarily lend support to a level of 75 µeq/L for a 3-5 year average, since it can decades or longer to reach the long term level.

p. 7-52. The ‘framing’ of this text is from the perspective of decreasing marginal protection as the level increases. However, conversely, this could be framed in terms of increasing certainty of protection as the level increases. Or, as the level is lowered, increasing certainty of loss of protection. The framing is quite important because it can bias the interpretation of the technical information. To avoid framing bias, all of these interpretations should be mentioned.

p. 7-53. It would be helpful to explain how CMAQ will be used operationally. For example, in order to have a 3-5 year average, consecutive annual CMAQ runs will be needed. The most recent averaging period can only be evaluated based on the most recently available CMAQ run, which may be one to three calendar years earlier than the current calendar year. Thus, non-attainment decisions would have a lag period of one to three years. Some discussion of this would be helpful.

p. 7-54. It would be helpful to have a table of all of the values that are input to F1, F2, F3, and F4, and of the values of F1, F2 (for those Fi derived from more than one input) associated with the ecoregions listed in Tables 7-5.1

### **Uncertainty/Sensitivity/Variability:**

#### **Appendix F**

p. F-3: ‘bootstrapping Monte Carlo type analysis’ – this term is very unclear. There are specific types of bootstrap and specific types of Monte Carlo analysis. A broader point, however, is that the statement on lines 13-17 seems to be contradicted by the analysis given in Appendix G. The

latter is an uncertainty analysis of AAI except that it is inverted such that combinations of the SO<sub>x</sub> and NO<sub>y</sub> concentrations are inferred. This paragraph should be rewritten for clarity.

p. F-13, table F-4.2. The header should be more clear that it refers to Annual Mean Bias Statistics and that it is based on CASTNET sites in the Eastern Domain.

p. F-20. Why is there not a similar graph for SO<sub>2</sub>?

p. F-21: the data in these graphs are not interpreted in the text.

p. F-22. There is essentially no citation or interpretation of data in the associated figures.

p. F-23. Define the box plots – mean? 95 percent frequency range? Since these are hourly data, why/how is this relevant to looking at annual averages or 3-5 year averages?

p. F-26. Are the data in the figure for 2002?

p. F-34, Figure F-15: define the meaning of the box and whiskers. Also for next page.

p. F-37. This comparison is not very informative, given that it is not on a consistent basis. The text could be more clear as to whether this comparison is really useful, other than just getting an idea of the typical magnitude of these transference ratios. It is not clear to the reader that these data can provide “extremely valuable diagnostic information.” What is “extremely” valuable about it? How can it be used to improve “CMAQ deposition processes”?

p. F-45: when translating “error in individual measurements” to regional values, one must pay careful attention to the spatial scale and averaging time.

What is a “bootstrapping analysis of the parameters”? This is never explained adequately. There are many kinds of bootstrap. The basic input data are not clearly presented or described. The use of the term “parameter” here is sloppy.

## **Appendix G**

The material in Appendix G is only useful if the reader can figure out what were there key inputs and the methodology used, neither of which are adequately addressed. For example, to merely describe the methodology as “bootstrapping” is not useful – there are many kinds of bootstrapping. What exactly was bootstrapped and how? The key input data should be clearly introduced and either presented or at least described or visualized if possible. The methods by which the data were bootstrapped should be clearly explained. The approach for Monte Carlo analysis and by which the probabilistic trade-off curves for SO<sub>x</sub> and NO<sub>x</sub> were derived must be adequately explained, including documentation of the key equation(s), key input distributions, analysis/simulation methods, and results. A rationale for using the mean values as the basis for the standard should be given. Some explanation should be given as to the implications of using upper or lower percentiles of the trade-off curve.

p. G-1: is there any statistical dependence among Neco, NH<sub>x</sub>, Tnoy, and Tsox? In what way is Neco from CMAQ?

There needs to be a table that clearly documents or explains the inputs to the ‘bootstrapping’ analysis, and associated figures that show the data.

How exactly were ‘these averaged values’ ‘bootstrapped using uncertainty in CMAQ modeling to obtain estimated uncertainty in the average values?’ What ‘uncertainty’ was used in CMAQ and how? What data were bootstrapped, and what kind of bootstrapping was done? For what averaging time? Geographic extent?

p. G-2: Results are shown but the input and method are not documented or adequately explained. For what averaging time? Annual averages? 3-5 year averages? Does “uncertainty” refer to a 95 percent probability range?

“Nleach was allowed to have 30% uncertainty.” (underline added). What is meant by “allowed”? Is this a judgment? Based on what? What type of distribution was assumed and why?

p. G-3: Figure G-2 – for what averaging time?

“Uncertainty in CMAQ values was included randomly...” what does this mean? How was this done? On what basis?

The text on the last few lines should be discussed more fully. What is the implication if the correlation is not zero? Is it possible to a sensitivity analysis to assess the results with 100% correlation?

p. G-4: is the ‘uncertainty’ range for a 95% probability range? What is the vertical line in each figure panel? What averaging time is used here?

p. G-5 “with the same uncertainty of 100%” – this is unclear.

p. G-6: unclear as to how uncertainty is “bootstrapped” if it includes arbitrary ranges of uncertainty. Bootstrap typically implies reference to empirical data, either by resampling or by use of a parametric model fit to data. Bootstrap does not usually describe sampling from arbitrary distributions. What was resampled here? What is meant by and what is the basis for ‘including’ 70 % uncertainty in BC and 5% uncertainty in Qo?

p. G-7: what is meant by “raw data” for these ecoregions? “The curve was then derived again using values from the derived distributions of...” this is not clear. There should be a basic equation or flow diagram to describe what was done.

In what way was Neco treated separately from the CL selection? This is not clear.

p. G-8. The two figures given here are not interpretable given the lack of clarity regarding input data and methods used. For example, if these results are based on averaging times of less than 3-5 years, then they would overestimate the uncertainty in the trade-off curves.

**Dr. Paul Hanson**

The summary of the available science is generally acceptable. I agree with the retention of the existing standard for protection human welfare from gaseous forms of nitrogen and sulfur oxides, and with the pursuit of a secondary standard based on ecosystems effects driven by total N and S deposition through the processes of acidification and nutrient enrichment. The focus on the development of a standard based on aquatic ecosystems seems appropriate and mandatory given the limited nature of data and defined response relationships for terrestrial ecosystems.

I am not providing a list of specific comments on this Policy Assessment document, because it is in final form and will presumably not be changed. I do, however, want to comment on word choices in the Executive Summary to highlight a remaining concern. That concern is that the document (in places) continues to suggest that N and S deposition is an ever present and current danger to all ecosystems. The Policy Assessment (PA) document, the REA and the ISA before it support a conclusion that current levels of N and S deposition represent a potential adverse impact on sensitive ecosystems through a range of mechanisms. This version of the PA includes improved language from the previous draft, but occasionally falls into the trap of suggesting that N and S deposition are bad for all ecosystems.

## Dr. Rudolph Husar

*Comments are in italics.*

### General

*Overall, this version of the PA document constitutes another significant improvement of this ground breaking regulatory document. The presentation of the proposed regulatory approach is much clearer and the document is much better structured and sections homogenized. This PA version shows strong responsiveness to past CASAC inputs. The content and structural improvements along with the cosmetic changes make this PA document nearly complete.*

*I recognize that this the 11<sup>th</sup> hour for the PA. Nevertheless below are comments and some remaining questions, including some persistent ones, for consideration by EPA.*

### Indicators for the Standard:

NO<sub>y</sub> - Total reactive oxidized nitrogen, NO<sub>y</sub>, as the ambient air indicator for oxides of nitrogen.

*The justification for the selection of atmospheric NO<sub>y</sub> as the oxidized N deposition indicator is not very compelling. HNO<sub>3</sub> gas dry deposition and NO<sub>3</sub> wet deposition contribute the overwhelming majority of oxidized N deposition. Both are measurable and measured parameters, why not use these as indicators for ox. N dep? The fact that NO<sub>y</sub> can be measured does not mean that it is a more robust indicator than HNO<sub>3</sub> gas and NO<sub>3</sub> wet dep. For receptors near the source, the effective multi-component 'transmittance function' is much different from the far source and applying CMAQ to properly account for these differences may be of tenuous robustness.*

*If there are other reasons for NO<sub>y</sub> as the indicator, those should be explained more clearly.*

SO<sub>x</sub> - Sum of gaseous sulfur dioxide (SO<sub>2</sub>) and particulate sulfate (SO<sub>4</sub>) as the ambient air indicator for oxides of sulfur.

OK

### Form of the Standard, AAI:

The *form* of such a multi-pollutant, deposition-related standard, should be ecologically relevant and include link ambient air indicators for oxides of nitrogen and sulfur, the related deposition of nitrogen and sulfur, and the associated aquatic acidification effects in terms of the ecological indicator ANC.

Aquatic acidification index (AAI), using a *simple* (??) equation to calculate an AAI value in terms of the ambient air indicators NO<sub>y</sub> and SO<sub>x</sub> and the relevant ecological and atmospheric factors that modify the relationships between the ambient air indicators and ANC.

$$AAI = F1 [units\ deposition?] - (F2[g/m^2,s] + F3[m/s]*(NO_y) [g/m^3] + F4[m/s]*(SO_x)[g/m^3])$$

*The equation may be simple but the complications are embedded in the factors F1 to F4. Also, these factors rely heavily on model estimates, so these 'simple' terms in AAI arise from very complicated calculations and they are also the carriers of much of the uncertainty in AAI determination. So, I would remove the word 'simple'.*

#### F<sub>1</sub> - Ecosystem Neutralizing Capacity

F<sub>1</sub> - Ecosystem's natural ability to provide acid neutralizing capacity and to neutralize nitrogen deposition through plant uptake and other processes; F<sub>1</sub> (as the other F factors) would be defined for each ecoregion by specifying ecoregion-specific values for each factor based on monitored or modeled data that are representative of each ecoregion. The AAI is to be evaluated over ecologically relevant regions, defined by Omernik Ecoregions, level III, 84. Factors F<sub>1</sub> through F<sub>4</sub> would be defined for each ecoregion by specifying ecoregion-specific values for each factor based on monitored or modeled data that are representative of each ecoregion.

*Is this another novel aspect of this new standard (?): Defining the areas of compliance for regions (not points) and determining non-compliance by regionally 'representative' values of all AAI terms...or just the F factors? While the NO<sub>y</sub> and SO<sub>x</sub> observations would be the anchors to reality but it would be the CMAQ that (1) estimates the F<sub>3</sub> and F<sub>4</sub> factors...and (2) spatially extrapolates these observations for each ecoregion.*

*This is a rather heavy burden for a model to carry. Could it happen that at the 'representative' observation site the location is compliant but the region is not compliant due to spatial gradients of any of the AAI terms? Can CMAQ withstand the scrutiny of the standard implementation process?*

#### F<sub>2</sub> - Acidifying deposition associated with reduced forms of nitrogen, NH<sub>x</sub>;

*So ammonia deposition estimate should be (1) entirely based on CMAQ, (or local model)? Why not use measured ammonia wet deposition data and model only the 'transference ratios'. Why would model NH<sub>x</sub> wet deposition be more robust than the observations? See below re data assimilation.*

F<sub>3</sub> and F<sub>4</sub>: Transference ratios convert NO<sub>y</sub> and SO<sub>x</sub> conc. to deposition

Specifying the values for the transference ratios based on CMAQ modeling results alone is preferred to an alternative approach that combines CMAQ model estimates with observational data

*The value judgment by EPA that CMAQ model estimates of the transference ratios in AAI is preferred to combined model-obs values is not shared by this reviewer. Yes, assimilating observations into atmospheric models is an inconvenience for the current models. However, regulatory arm of EPA could and should press its research arm to pursue such CAMQ data assimilation upgrade, particularly for wet deposition. Such a assimilation-enhanced CMAQ*

*could benefit many applications of the official EPA regional model beyond this secondary standard.*

*Since the units of F3 and F4 transference ratios are m/s, I still do not follow why these mysterious F factors could not be called effective [or total wet+dry] deposition velocities. By definition, the deposition velocity is the ratio of ambient concentration and deposition.*  
Averaging Time for the Standard

All calculated as annual average values, whether based on water quality and hydrology data or on CMAQ model simulations. The same annual averaging time for the ambient air indicators as is used for the factors in the AAI equation.

Annual average AAI values over 3 to 5 years to provide reasonable stability in the resulting index value, in light of the relatively high interannual variability expected.

*Agree, annual arithmetic average of atmospheric, deposition and ecological variables. No further comments now.*

Level of the Standard

The *level* of a standard based on the above indicators, alternative forms, and averaging times should be within the range of 20 to 75  $\mu\text{eq/L}$ .

*If Aquatic Acidification Index (AAI) is the standard and its units are  $\mu\text{eq/L}$ , then how does one combine the F1 [ $\mu\text{eq/L}$ ] terms with the deposition terms F2, F3, F4 [ $\text{g/m}^2, \text{s}$  or  $\text{meq/m}^2\text{-yr}$ ]?*

## Dr. Dale W. Johnson

The primary topic assigned to me for review is “Levels” within Chapter 7, so I will first address that segment, then a few additional comments on other sections below.

### Chapter 7 LEVEL

As I indicated in my last review, I see no problem with the ANC target levels chosen. Also as indicated in my last review, I do think that more attention should be given to capacity vs intensity effects of acidification. I have raised this several times, but still see some confusion, so I will take the liberty of providing an old paper and some visuals I use in class to illustrate this point. Figure 1 depicts the situation that most people think of when considering acidification, namely, that the soil acidifies and the waters passing through it (soil solution) also acidify as a result. This mechanism, which John Reuss (Reuss and Johnson, called the capacity effect, usually takes a long time, perhaps decades or centuries, and presumes that the soil begins in a non-acidic, or at least not extremely acidic, condition. Figure 2 illustrates a second mechanism by which soil solution can acidify: the soil is naturally acidic, but under pristine conditions, the soil solution is not necessarily acidic because the concentrations of strong acid anions ( $\text{Cl}^-$ ,  $\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$ ) are not high enough to balance a very high concentration of  $\text{H}^+$  or  $\text{Al}^{3+}$ , which are the two major exchangeable cations on the soil in this case. When strong acid anions are introduced to this soil, however, acidification of the soil solution is instantaneous, as the incoming cations associated with these anions (whether acidic or basic) are exchanged with the soil and the soil releases  $\text{H}^+$  and  $\text{Al}^{3+}$  from the exchange sites into soil solution. The second mechanism is well-known in soil science in that soil paste pH measured in 0.01 M  $\text{CaCl}_2$  is nearly always higher than when it is measured in water for exactly the same reasons (both methods of measuring soil pH are commonly employed). It is important to note that **no change in the soil is required for the second mechanism**. It is also important to note that **acidic soils are a necessary but not sufficient condition for the acidification of soil solution**. You may wonder if the second statement is really true; I attach a very old paper of mine which shows that acidic soils in Costa Rica (pH 4.0 to 4.9 in  $\text{H}_2\text{O}$ ; see Table 2) produce soil solution pH values around 6 (Figure 4). In the Alaskan site, soil pH ranges from 3.95 to 4.74, but in this case, soil solutions are acidic in forest floor and somewhat less so in the A2 horizon because of the presence of organic acids, but soil solution pH increases as the organic acids are precipitated out in the B2ir horizon (these are Spodosols).

The differences in these two mechanisms are very important. In mechanism 1, acidification takes a long time to occur. In mechanism 2, however, acidification is instantaneous as soon as strong acid anions added to the system. Since there is no reason to believe that soils in humid regions spontaneously alkalize (desert soils do, but humid region soils spontaneously acidify, not alkalize), recovery from mechanism 1 is either slow or non-existent. Recovery in mechanism 2, however, closely tracks strong acid anion deposition (as we seem to be seeing currently in the Adirondack lakes, for example). John Reuss provided a succinct discussion of capacity and intensity effects on pp 71-72 in Reuss and Johnson (1986).

I hope this brief primer from my class notes does not come across as condescending, it certainly is not meant to be so; my intent is to illustrate what I have been talking about. This of course

only applies to the scenario where waters flow through soil before entering lakes or streams, not surface flow or other mechanisms by which atmospheric deposition might enter aquatic systems more directly.

Given the above, I have problems with the statement on p. 7-49, for example, which states categorically that “Ecosystems become adversely impacted by acidifying deposition over long periods of time...”. The next paragraph on p. 7-49 is all about timing of acidification, and I really think that the above discussion is highly relevant to it.

Other Comments:

p. 3-22, first whole paragraph: Finally we agree on how to state this issue of potential benefits of N deposition!

p. 3-25: I would add a segment on unintended consequences for C balance issues. Sensing great reluctance to do this on the part of the authors, however, I do not require it; I simply say that if I were an author, I would add it.

p. 7-19: As I have indicated in previous reviews, this model seems to assume that there will be no response in base cation concentrations to inputs of strong acid anions. Am I wrong? If not, this flies in the face of experiences in nearly all artificial acidification experiments, not to mention what happens to soils when they acidify (capacity effect). For mechanism 2 above, this approximation is more realistic, but not in the cases where soils are assumed to be acidifying (mechanism 1).

p. 7-43: The discussion of capacity and intensity effects above is relevant here.

Reference:

Reuss, J.O., and D.W. Johnson. 1986. *Acid Deposition and the Acidification of Soil and Water*. Ecological Studies No. 59. Springer-Verlag, New York. 118 p.

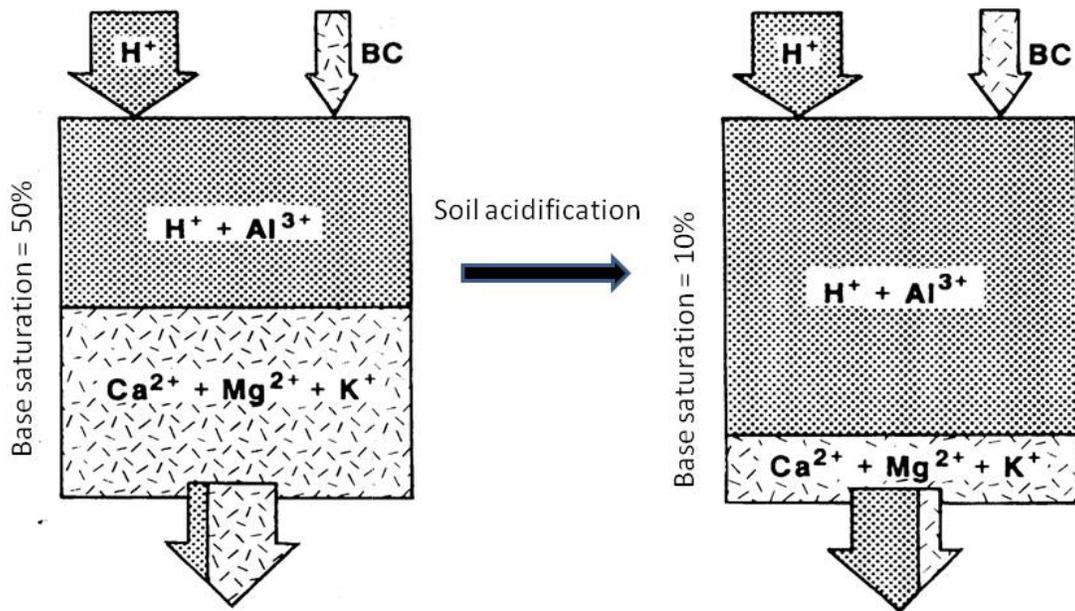


Figure 1. Schematic diagram of the capacity effect. In this case, the soil is subjected to high rates of acid deposition. At first, most of the incoming acid is exchanged for base cations (BC), and the soil solution is not acidic. As the soil acidifies over time, a greater proportion of cations on the soil exchange sites are acidic cations and they therefore dominate soil solution as well.

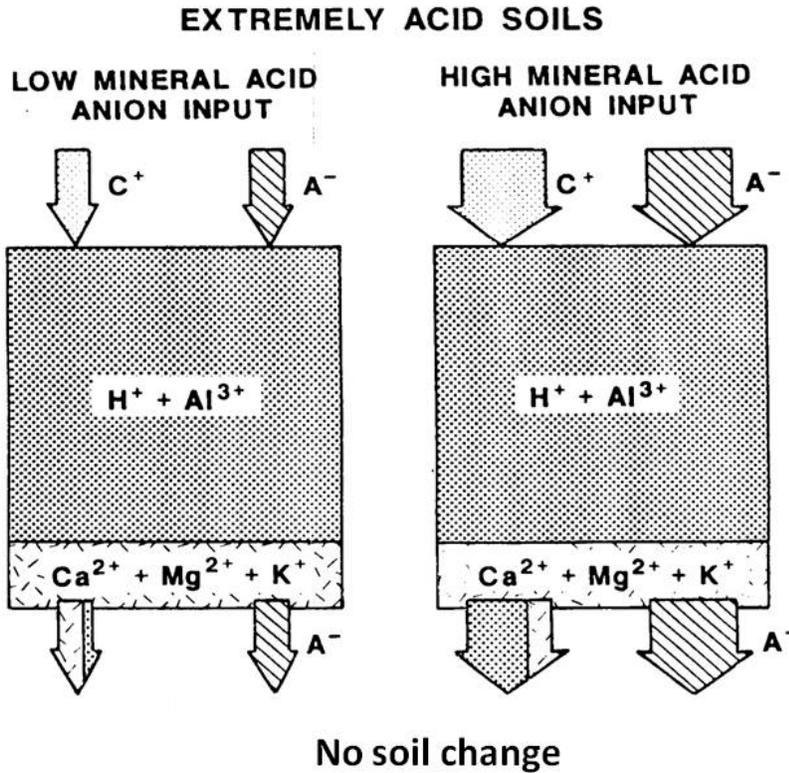


Figure 2. Schematic diagram of the intensity effect. In this case, the soil already naturally acidic. Under low strong acid anion deposition conditions, only low concentrations of acidic cations appear in soil solution because soil solution strong acid anion concentrations are low. With increased strong acid anion deposition and leaching through the soil, charge balance requires that soil solution total cation concentrations increase, and since the soil is acidic, exchange equations predict that the acidic cations will constitute a major proportion of this soil solution cation concentration increase. Soil solution acidity can increase or decrease very rapidly depending on strong acid anion inputs. No change in the soil exchanger itself is required.

## Dr. Naresh Kumar

This version of the Policy Assessment Document (PAD) for secondary standards of SO<sub>x</sub> and NO<sub>x</sub> is greatly improved in its presentation of key concepts, although many key issues still remain unaddressed. The specific comments on different elements of the standard are:

### Indicators

The use of ANC as an ecological indicator for aquatic acidification is not fully supported. EPA recognizes that ANC is not itself the causative or toxic agent for adverse aquatic acidification effects, it proposes to use ANC as the indicator because it is associated with the causative agent, pH. However, the empirical data suggests that this relationship is not universal because of the broad distribution of dissolved organic carbon (DOC) and carbonates in real water bodies. Just because ANC is a more reliable indicator from a modeling perspective is not a good enough reason because convenience shouldn't trump science when it comes to establishing national level standards. The empirical evidence shown by EPA to show relationship between adverse effects and ANC is derived from two regions of the country – Adirondacks and Shenandoah – and is not universal. EPA claims that ANC-deposition link is superior to the pH-deposition link because the relationship between pH and deposition is extremely nonlinear and complex. However, given that pH is a direct causative agent for adverse aquatic effects, EPA should have explored using pH as the aquatic indicator rather than completely dismissing it in favor of ANC

### Form

The form of the proposed standard uses the aquatic acidification index (AAI) that links ambient indicators to deposition metrics. The major concern with using this form of the standard is that most of the parameters in the AAI equation are determined using models that have not been adequately evaluated. The specific comments are:

1. **Lack of adequate model performance evaluation** – Given that this is the first such use of models in establishing standards (rather than the use of monitored values); the burden on validating the used models has to be much higher than when they are used in a relative sense (e.g., during implementation of the standards). However, EPA has continued to ignore previous requests from the CASAC panel to conduct a comprehensive model performance for CMAQ. The performance statistics for the major species shown in Table F-2 are inappropriate and misleading because they show average biases over the whole country for one whole year. What one needs to know is how much the model may be biased at the ecoregion level, and the PAD has failed to do that. Even when showing monthly time-series plots, EPA uses domain-wide averages rather than show the time-series at individual characteristic sites, as is the norm. EPA's approach can be misleading because it can mask the true error by combining regions of over-predictions with regions of under-predictions (often termed as compensation of errors), and is unacceptable when EPA is proposing to use these models in establishing national standards. Independent model performance evaluations of CMAQ conducted by EPRI (and communicated to the CASAC panel) has shown that regional biases in predictions of species of concern (SO<sub>2</sub>, SO<sub>4</sub>, NO<sub>3</sub>, NH<sub>4</sub>, wet deposition) can range from -100% to +200%; something that is

completely masked by EPA's approach. The request to EPA from the CASAC members to conduct comprehensive evaluation has been made for each version of the PAD, REA and ISA, thus EPA has had enough time to conduct such evaluation, but it has failed to do so. Given the lack of a complete evaluation of CMAQ derived values with measurements and the lack of usefulness of the limited model performance that is shown, no degree of confidence can be placed in the use of the models to generate the desired parameters in the AAI equation.

EPA has used the PRISM model to apply adjustments to wet deposition based on precipitation data. However, EPA has not shown how this selective application of bias adjustment affects the overall mass conservation. This also contradicts EPA's assertion of not combining measured and modeled data in development of the transference ratios.

The lack of robust model performance is also evident for the watershed models. In the appendix, EPA provides comparison of the MAGIC model results to calibration data; by definition, these align well because the model was calibrated to fit these data. EPA notes that the performance outside of the calibration period is not as good and refers to the REA document. However, the REA document shows model performance for two Adirondack lakes and two Shenandoah streams from a population of 104 waterbodies in the combined dataset. Unless EPA also shows the model performance for the remaining 100 waterbodies, it is difficult to draw any conclusions on the adequacy of the model performance. EPA has failed to provide any reasons for not showing the model performance for all the waterbodies for which it conducted the analysis. A bigger concern may be the bias introduced by the use of the MAGIC model or the F-factor approach in calculating the critical loads. Evaluation of the MAGIC model at the 44 lakes in Adirondacks, as shown by EPRI in its comments to CASAC on the second draft of the PAD, indicated that the MAGIC model resulted in a "sum-of-strong-bases to sum-of-strong-anions ratio" (tantamount to the F-factor) of 0.42 whereas the real data show a value of 0.87 (this would imply the critical load calculations using the MAGIC model would be biased low). EPA has shown that the F-factor approach gives critical loads that are even lower than the MAGIC model. Thus, no matter which approach is used by EPA (because of data limitations, EPA will most likely rely on the F-factor approach), the critical loads calculated will be biased low. This can have major implications on the level of the standards chosen.

- 2. Stability of transference ratios** – A major concern with the transference ratios is that although they are the critical links between NO<sub>y</sub> and SO<sub>x</sub> ambient concentrations and their deposition, they are derived using a model that has not been adequately evaluated. EPA has shown the year-to-year change in transference ratios using the same model to evaluate its stability. However, the bigger concern is not the stability from year-to-year, but the variability in the ratios as determined from different models. EPA agrees that transference ratios constructed from different modeling platforms do exhibit significant differences; however those results are not shown. The fact that the choice of a model is so critical in determining the link between the ambient indicators (NO<sub>y</sub> and SO<sub>x</sub> ambient concentrations) and deposition, it further affirms why an adequate model performance needs to be part of the process. EPA contends, "the use of a modeled constructed

transference ratio is based on the same premise by which we use models to estimate deposition in the first place”. However, two wrongs don’t make it right. It is wrong to use a model without adequate performance evaluation to estimate deposition when establishing a national standard, and it doubly wrong to use that as a reason to endorse the use of the same model for establishing a critical link in the form of the standard (the AAI).

3. **Spatial Aggregation** – EPA is proposing to use ecoregion Level III as spatial areas over which values for the factors in the AAI equation are quantified. There are a total of 84 such regions in the contiguous United States. One problem with using such large spatial regions to define the standard is that one has to use averaged quantities over regions that can be quite inhomogeneous and disparate. A more critical issue is whether use of these ecoregions may introduce biases in calculation of critical loads. Water bodies that are actively sampled in the U.S. are most often predominantly located in the more acid sensitive regions. Using these sampled data to calculate critical loads for the whole ecoregion will lead to critical loads that are biased low. The representativeness of available data to use for an ecoregion is a critical issue that has not been addressed by EPA. The biases resulting from use of currently available water chemistry data would imply that a given percentile of the distribution of estimated critical loads for surface waters would be protective of a higher percentage of surface waters in most regions because of un-representativeness of the data. For example, critical load calculated based on 70<sup>th</sup> percentile of waterbodies for which data are available will be lower than the critical load calculated based on 70<sup>th</sup> percentile of all waterbodies in the whole ecoregion because of skewness of data towards more acid-sensitive waterbodies.

## Level

EPA suggests a level of the standard in the range of 20-75  $\mu\text{eq L}^{-1}$  for the ecological indicator, ANC. Rather than choosing a number arbitrarily in this recommended range, we should consider all the factors that are critical in linking adverse aquatic effects to causative agents and also examine the factors that may be causing biases in EPA’s approach before deciding on the ANC level for the standard. EPA has rightfully stated that pH is a better causative link with adverse aquatic effects, so one should use studies linking pH to adverse effects when determining an appropriate level. Table 3-1 on Page 3-8 shows different studies linking fish mortality response to pH. The range of pH values causing fish mortality is between 4.5 and 6 for various studies except one study where the pH was at 6.5. Assuming a conservative value of 6.5 for pH that causes adverse effects, one can determine that the corresponding ANC (from Figure 7-2) is about 25  $\mu\text{eq L}^{-1}$ . This would be a better approach than relying on a single study linking ANC with adverse effects (Figure 7-4), which shows increasing effects from 0 to 100  $\mu\text{eq L}^{-1}$ , but also shows increasing uncertainty above 20  $\mu\text{eq L}^{-1}$ . In regions where ANC is low, but inorganic aluminum levels are also low, the fish populations have been shown to be thriving, so one shouldn’t rely only on a single study. Another study in western Virginia showed that at ANC above 50  $\mu\text{eq L}^{-1}$ , the brook trout populations were not affected.

Another factor that is important is that as shown previously, EPA’s approach would lead to critical load values that would be biased low because of the aggregation at the ecoregion level

and because of the use of the MAGIC model and the F-factor approach. This would in turn cause the NO<sub>y</sub> and SO<sub>x</sub> allowable levels to be biased low. Thus, in the absence of any other criteria, selection of the ANC level should be conservative towards the low end of the recommended range of 20-75 µeq L<sup>-1</sup> to balance against the biases introduced by the EPA approach. Moreover, in this case the evidence suggests (as explained previously) that the appropriate ANC value in absence of any bias should be about 25 µeq L<sup>-1</sup>. However, given the other biases, one can justify 20 µeq L<sup>-1</sup> as the appropriate ANC value for the standard.

The choice of the percentile of waterbodies to be protected in an ecoregion is also an important part of the standard. EPA has recommended a range of 70<sup>th</sup> percentile to 90<sup>th</sup> percentile of waterbodies to be protected in ecoregions classified as acid-sensitive. However, given the fact that most of the data for estimation of critical loads is predominantly available at waterbodies in acid-sensitive regions, these percentile levels could lead to an extremely strict standard. Consider a thought experiment where an ecoregion has 1000 waterbodies of similar sizes and assume that 200 of those waterbodies are acid-sensitive. Now assume that a total of 100 waterbodies in this ecoregion have water chemistry data with 80 of those waterbodies to be acid-sensitive and the other 20 to be non acid-sensitive. These numbers are not out of ordinary and may be quite realistic. A critical load based on 70<sup>th</sup> percentile of these 100 waterbodies (for which data are available) would represent an extremely acid-sensitive waterbody and may be overly protective of all the acid-sensitive waterbodies in that region, notwithstanding the large number of non acid-sensitive waterbodies. A percentile level of 70 or larger is appropriate only if majority of the waterbodies in an ecoregion are acid-sensitive or if the data are available in equal proportion for both acid-sensitive and non acid-sensitive water bodies. Given that this is not the case, a 50<sup>th</sup> percentile may be a more appropriate level, although still conservative because of the skewness in availability of data.

### **Uncertainty Analysis**

It is difficult to judge the adequacy of the uncertainty analysis performed by EPA because of lack of details. The purpose of the uncertainty analysis as requested by CASAC was to estimate the degree of confidence in the response surfaces of NO<sub>y</sub> and SO<sub>x</sub> given the high degree of uncertainty in the individual terms of the AAI. The way the EPA has conducted the uncertainty analysis using bootstrapping, the uncertainties in individual terms may have been minimized. For example, EPA assumed an uncertainty analysis of 100% for Ndep at the grid level, but averaged over the ecoregions the bootstrapped distribution resulted in an uncertainty of only 10% and 15% for the two ecoregions. It seems the law of large numbers is at play here, i.e., when averaged over a large number (because of the large ecoregions) the average distribution will be closer to the expected value. Thus, EPA's approach of using bootstrapping seems questionable. By focusing on the uncertainty of the average parameters, EPA has ignored the variability of the values in the ecoregion, which is a critical omission because variability also affects the degree of confidence in a given value of a parameter.

Instead of using the mean values and the questionable bootstrapping method to assign uncertainty, EPA should have used the distribution of the actual individual parameters in an ecoregion. If the distribution in an ecoregion is homogeneous, the effect of variability would be minimal. Under those conditions, the degree of confidence in the response surfaces for SO<sub>x</sub> and

NO<sub>y</sub> is influenced solely by the uncertainty in the measurements or estimations of the AAI parameters. EPA's uncertainty analysis ignores the impact of variability in judging the degree of confidence in the response surfaces by assuming average conditions and defining fixed levels of uncertainty for the average value of the parameters, and thus doesn't serve the purpose as intended by the CASAC. Thus, the uncertainty analysis performed by EPA doesn't provide any insights into the confidence level of the standard because of the lack of clarity and the arbitrariness of the assumptions used. It is almost as good or as bad as the qualitative uncertainty analysis performed earlier and repeated in this version of the PAD. Both these analyses (qualitative and quantitative) purport to undermine the actual uncertainty in the system.

## Dr. Myron Mitchell

Comments on “Indicators” in Chapter 7 entitled “CONSIDERATION OF ALTERNATIVE STANDARDS FOR AQUATIC ACIDIFICATION”.

### General Comments

The introduction to this chapter (p. 7-1 through 7-3) does a good job of giving an overview of the overall approach and its rationale. Throughout this Chapter and the entire document the term “species is used extensively”. I would suggest that in future documents for those places when the “species” refers to “chemical species” that this term be provided explicitly so as to avoid any confusion with “biological species”. In the Panel’s previous discussions it was strongly suggested that the importance of ammonium and ammonia atmospheric deposition be incorporated into the standard. The beginning of this chapter (Section 7.1.2) suggests that only the NO<sub>y</sub> chemical species be utilized in the standard, but later (pages 7-21 through 7-23) the importance for incorporating NH<sub>x</sub> deposition is included. More consistency in the Chapter would have been helpful. In some of the discussions of this section the consistency of the arguments for the use of individual nitrogen species versus NO<sub>y</sub> chemical species could have been improved. However, it is indicated (page 7-9) that “The index also accounts for the contribution of reduced nitrogen to acidification”. In general the Chapter provides important arguments that link the aquatic acidification index (AAI) to ANC (Acid Neutralizing Capacity) and the use of ANC as important parameter for evaluating the effects of S and N deposition. I am still concerned with respect to the discussion of how internal S sources are not treated in the model formulation:

$$CL_{ANClim}(N + S) = ([BC]_{0*} - [ANClim])Q + Neco \quad (7-2)$$

as well as the version to be used in the actual Aquatic Acidification Index (AAI):

$$AAI = F1 - F2 - F3[NO_y] - F4[SO_x] \quad (7-12)$$

where

$$F1 = ANClim + CL_r/Q_r;$$

$$F2 = NH_x/Q_r = NH_x \text{ deposition divided by } Q_r;$$

$$F3 = T_{NO_y}/Q_r; T_{NO_y} \text{ is the transference ratio that converts deposition of } NO_y \text{ to ambient air concentrations of } NO_y; \text{ and}$$

$$F4 = T_{SO_x}/Q_r; T_{SO_x} \text{ is the transference ratio that converts deposition of } SO_x \text{ to ambient air concentrations of } SO_x.$$

It is indicated in the text (page 7-20) that more details are provided in Appendix B, but I do not see any treatment associated with internal S sources. There is no consideration in the appendix of the potential importance of organic S mineralization, sulfate desorption or the weathering of sulfur minerals. The assumption of the sulfur content in these ecosystems to be in steady state is not correct. In future efforts the importance of this internal S source in affecting ecosystem response to atmospheric deposition needs to further consideration by EPA.

There is extensive discussion related to the rationale for using “transference ratios” versus “deposition velocities (pages 7-23 through 7-26). Some of this discussion does not match well with the introductory aspects of the chapter that suggest the importance of using deposition velocities of individual chemical species (e.g., pages 7-5 through 7-7). There is considerable argument provided on the use of CMAQ to develop transference ratios (pages 7-23 through 7-26). The selection of the Omernik’s ecoregion classification for geographical separation is well justified and builds upon previous efforts by EPA. The selection of ecoregion level III resolution seems appropriate although it will certainly be challenging to provide parameters for the 84 defined regions. The further delineation of two broad groupings of ecoregions (acid sensitive and relatively non-acid sensitive) is a useful approach in moving this effort forward. It was also helpful to identify those ecoregions with insufficient data needed and to use median values of  $CL_r$  and  $Q_r$  from all the relatively non-acid sensitive ecoregions.

Although I agree that for the current standards the appropriate focus is on chronic acidification (annual averages) as discussed in Section 7-3, there should be some statement that indicates that with climate change resulting in greater amplitudes in hydrological events that episodic acidification may become a more dominant and important factor in the future. The discussion on the need to focus on a range of ANC values is useful, but it is not completely clear why the final selection of the range was from 25 to 75  $\mu\text{eq L}^{-1}$  versus 0 to 100  $\mu\text{eq L}^{-1}$  (pages 7-46 through 7-47). However, some later arguments (e.g., pages 7-50 through 7-51) including what standards have been set by others helps to justify the former range.

Section 7.5 on “Considerations associated with alternative standards” provides some useful discussion on the applications of various standards and how their utilization affects attainment among the various ecoregions. The discussion is useful on how the implementation of the standard is affected within the target ANC range of 20 to 75  $\mu\text{eq/L}$  and within representative percentile values within the range of the 70th to 90th percentile identified in Section 7.2.5.3 with higher percentiles corresponding to a lower critical loads. The overall findings as outlined in this section provide supportive information on the implementation of the AAI approach with results being consistent with known spatial and temporal factors associated with surface water acidification and potential for recovery. Will the implementation of this approach and resultant standard have any support for those geographic regions that are more concerned with N enrichment in affecting ecosystem structure and function versus acidification?

Table 7-6-1 that provides a “Summary of Qualitative Uncertainty Analysis of Key Components of the AAI” is very important”. I am not sure that all of the uncertainty associated with the model formulations is clearly articulated including current and future uncertainty about the estimates of Neco. There is no indication in the table of any implications associated with the assumption that the sulfur inputs and outputs are not in steady state.

The Summary Section (7.7) is a useful synthesis of the overall approach to this new standard. On page 7-77 it is stated that “consideration should be given to establishing a new ecologically relevant standard(s) to provide increased protection from deposition-related effects of oxides of nitrogen and sulfur on sensitive ecosystems”. Clearly this approach considers the combined effects of N and S deposition on acidification, but will the absence of nutrient effects

compromise the goal in the establishment of an “ecologically relevant standard”?

Throughout the document the term “neutralize nitrogen deposition” is used and its meaning may be confused. When this term is first used it should be defined as gaseous loss and N storage (e.g., page ES-8 and 7-18).

### Specific Comments (Changes are indicated *in italics*)

Page(s)	Comment
7-3	In discussing the effects of atmospheric deposition of N and S compounds it should be mentioned that not only is the effect associated with “acidification”, but also to the generation of mobile anions (e.g., $\text{SO}_4^{2-}$ and $\text{NO}_3^-$ ).
7-11	Change to “ <i>low</i> pH”.
7-11	Change to “ANC is a more <i>useful</i> indicator”.
7-11	Change to “acidification through the input of strong acid, <i>mobile</i> anions (e.g., $\text{NO}_3^-$ and $\text{SO}_4^{2-}$ ) that <i>may</i> shift”.
7-11	Change to “transformation through soil <i>S and N mineralization and</i> nitrification processes and subsequent drainage from terrestrial ecosystems”.
7-11	Change to “surface water pH; trivalent aluminum, $\text{Al}^{+3}$ , <i>concentrations;</i> ”.
7-12	Change to “Aluminum and <i>some</i> other metals <i>may be</i> causative toxic agents that directly impair biological functions”.
7-13	Change to “solution must satisfy the condition that <i>the sum of</i> negatively charged species must be balanced by <i>the sum of</i> positively charged species”.
7-15	Not sure what the meaning of the statement of “relationship can be extremely nonlinear” is trying to convey.
7-16	Change to “as temperature, <i>partial pressure of</i> $\text{CO}_2$ , mixing conditions of a water body, and the levels of other several chemical species in the system including $\text{HCO}_3^-$ ”.
7-17	Change to “since the onset of <i>marked increases in</i> anthropogenic nitrogen and sulfur deposition”.



## Mr. Rich Poirot

### Form

Given the multi-pollutant, multi-media, environmentally modified, geographically variable nature of SO<sub>x</sub>/NO<sub>x</sub>-related aquatic acidification effects, the form of any national ambient air standard intended to address those effects is guaranteed to be complex. The form of the NAAQS proposed in this final PA is unavoidably complex, but is also entirely consistent with and directly reflective of the current scientific understanding of the problem. Each element or range of options for the form presented in section 7.2 of the PA is logically derived, well justified and clearly explained. The high clarity of presentation, added specificity of details, and staff recommendations of options or ranges of options for individual elements of the form are all notable improvements from the previous draft PA, and are directly responsive to previous CASAC review comments.

The proposed form consists of two general parts - a chemical component, and a spatial component. The chemical component (AAI) links an environmental indicator (a protective level of ANC in surface waters) to measurable concentrations of SO<sub>x</sub> and NO<sub>y</sub> in the ambient air. This chemical component can be expressed in terms of the (7-12) equation:  $AAI = F1 - F2 - F3[NO_y] - F4[SO_x]$ . For any given spatial area and target ANC value, each of the “F” factors in this equation can be developed from pre-existing measurement and model data. For any specified level of the standard, compliance can then be directly determined from new and continuing measurements of SO<sub>x</sub> and NO<sub>y</sub>.

The proposed form also includes a spatial component which would define the spatial areas (Omernick’s Ecoregions, Level III), over which separate, spatially aggregated, chemical AAI calculations would apply. It divides those ecoregions into subsets considered “sensitive” and “non-sensitive” to acidification, and also proposes separate ranges of percentiles of critical loads for surface waters to be protected from acidification within each sensitivity category. The current staff recommendations include a using a more protective range of the 70<sup>th</sup> to 90<sup>th</sup> percentile critical load values in acid sensitive ecoregions and the 50<sup>th</sup> percentile critical load in non-sensitive areas (either averaged across individual areas or across all non-sensitive areas).

The proposed chemical component of the form appears to be technically sound and the AAI equation directly parallels the conceptual model of our scientific understanding of the problem. Since the F terms can be derived from existing data, the inherent complexities of the form could presumably be reduced to a nomograph or set of look-up tables, developed in advance for each ecoregion. It is notable that the important factors F2, F3 and F4 would be entirely derived from CMAQ model results. This partial reliance on CMAQ represents a significant departure from previous NAAQS proposals. This isn’t necessarily a problem, and there may be several benefits to combining models and measurements in setting this (or other future) NAAQS, as well as in the implementation phase. The accuracy of the factors supplied here by the model is also inherently difficult to evaluate (almost by definition, since if there were good measurements available to evaluate, the model results wouldn’t be needed). For a standard with a form like this to be effective, it will be important to continually scrutinize model performance, adding measurements where possible to help evaluate and improve model performance, and to publicly

expose all the relevant details of the model calculations (i.e. show us all the gridded concentration and deposition fields – not just the ecoregion AQI look-up tables).

The proposed spatial components of the form also generally appear to be sound, and the use of Omernick's ecoregions (Level III) seems like an appropriate and positive new development for secondary NAAQS intended to protect the environment – especially where there are geographical variations in the inherent sensitivity of ecosystems to pollutant effects. It is difficult to evaluate the logic or “wisdom” of the proposed percentiles (70% to 90%) for critical loads of lakes in sensitive ecoregions when taken in isolation. However, when these percentile ranges are combined with alternative levels within the staff recommended ANC range of 20 to 75  $\mu\text{eq/L}$ , the resulting range of combined levels and forms appears to generally focus on the right problem areas, and to indicate reasonable degrees of the problem severity (especially at the higher percentiles and levels). This “logical performance” of the combined proposals of level and form provides confidence in the overall design of the standard, in the individual elements of the level and form, and in the ranges recommended by staff. These combined recommendations provide the Administrator with a broad but reasonable range of adequate to minimally protective options for the standard.

The division of ecoregions into “sensitive” and “non-sensitive” subsets, with a more protective percentile applied to the sensitive areas, seems reasonable. It's not entirely clear, however, how the various sensitivity criteria will actually be applied. The naturally poorly buffered coastal plain areas (regions 8.5.4, 8.5.1, 8.3.5, and 8.5.3) are initially identified as sensitive areas, are included as such in Tables 7-5 and in the various appendices, and exhibit some of the most extreme potential exceedances at the 70<sup>th</sup> through 90<sup>th</sup> percentiles at all potential levels of the standard. But these areas are later identified as anomalous, and found in CMAQ model runs with future SO<sub>x</sub> & NO<sub>x</sub> emissions reductions to be “non-responsive” to SO<sub>x</sub> and NO<sub>x</sub> deposition changes. It is suggested (p. 7-63) that “responsiveness to deposition change” might be included as one of the acid sensitivity criteria, in which case these coastal plain areas would be designated non-sensitive. So are they in or out, and if they are out, is the lower 50<sup>th</sup> percentile sufficient to assure that they will not be identified as out of (or at least not way out of) compliance?

I think adding the “responsiveness” criteria to determine relative sensitivity is reasonable and appropriate, especially in a case like this where the scientific understanding of ecological response to changing atmospheric loading is reasonably well understood, can be further documented with observations from the recent past, and can be periodically evaluated by “ground truth” (surface water) data as the standard is implemented. In the event that subsections of ecoregions exhibit substantially greater (or lower) responsiveness than the region as a whole, consideration should be given to further dividing those regions. Establishing such subregions may also be desirable for regions which exhibit large spatial variability in T<sub>SO<sub>x</sub></sub>, or T<sub>NO<sub>y</sub></sub> ratios or SO<sub>x</sub>, NO<sub>y</sub> concentrations. I encourage the Agency to include “responsiveness to deposition change” in its initial screening criteria to determine relative “sensitivity”, and think it will also be a useful metric for determining and evaluating strategies in the implementation phase. Since determination of responsiveness, is like several other elements of this NAAQS, dependent on CMAQ model performance, it will be important to continually scrutinize model performance, adding measurements where possible to help evaluate and improve model performance, and to publicly expose all the relevant details of the model calculations.

**Mr. Dave Shaw**

## **General Comments**

I appreciate the scope and challenge of your task in establishing a NAAQS which will appropriately protect our ecosystems and linking that with ambient air concentrations. The form of the standard needs to be ecologically relevant and I believe that the form proposed in this latest version of the Policy Assessment (PA) is a rather good one, albeit with some considerations and concerns.

First and foremost, I do not see where the PA has addressed a concern which I have documented with each set of my previous comments. The concern is the lack of real monitored data. A clear commitment to the development of a relevant, reliable and consistent monitoring network needs to be shown. This should include a timeframe as well as expectations. By including a plan, and taking advantage of this multi-pollutant approach, we can make the best use of limited resources, for example, by using one monitoring location for several pollutants.

In addition to air monitoring, the PA needs to address what appears to be a lack of commitment to adequately measuring water quality indicators. If this is not addressed, the success or failure of the overall program will be indeterminate. The PA discusses the TIME and LTM programs, but this does not reach into each ecoregion. I recommend establishing a federally funded effort to measure water quality at 5-10 lakes in each ecoregion monthly starting as soon as possible. It would be advantageous to focus most of the water sampling on the moderately impacted lakes which are likely to show the most significant progress from changes in air quality over the timeframe of subsequent NAAQS reviews.

While the need for real data is clear, it is understandable that model predictions must be used in such a complex effort, it is unfortunate that so much of the F3 and F4 factors are based on model predictions. CMAQ is certainly well-tested and constantly being improved, but it has a vast array of options. For example, chemical mechanisms, physical packages (boundary layer dynamics, clouds, etc.), and initial/boundary conditions are only a few that you can get a range of answers for. States, MJO's, tribes, and other entities may want to use different, perhaps more recent, meteorological years, different emission inventories, which may give more reliable results, but different from other entities across the United States.

The use of the target ANC/critical load approach for surface waters has sound scientific footing and I feel is a proper method to move forward with for this multi-pollutant approach. The area that I have some concern with is the linkage which relates back to ambient air concentrations of  $\text{NO}_y$  and  $\text{SO}_x$ . Each of the four F factors, especially  $\text{NH}_x$  deposition (F2) and transference ratios (F3 and F4) have uncertainties and cannot be tested/verified. High quality representative air and water quality monitoring data are necessary in order to develop these modeled relationships. Again, this is why the program must have a strong emphasis on environmental monitoring.

I am focusing on data because model predications are based on collected data, so the model outcome can only be as good as the data which is used. If EPA decides to rely on model data over real data, model outcomes will become less reliable and as a result may become less

protective. On that note, it would be helpful to have some discussion comparing modeled estimates of transference ratios at co-located CASTNet/NADP sites where you have measured wet and estimated dry deposition.

Because this approach is so model dependent, some discussion on the response during a situation where the concentrations of  $\text{NO}_y$  and  $\text{SO}_x$  are within the allowable limits but the lakes still do not meet the critical ANC in X percent of the lakes/streams would be helpful. This scenario seems possible within ecoregions such as the Adirondacks where there is a lot of spatial variability between different lake classes.

I believe that the PA should be very clear about what the standard is and how progress will be assessed. Is the standard the AAI, the ambient air concentrations of  $\text{NO}_y$  and  $\text{SO}_x$ , or the ANC? Also, the PA needs to maintain a consistency in labeling tables and such with the word “standard.”

## **Dr. Kathleen Weathers**

This final version of the PA shows considerable improvement over the previous draft that the committee reviewed. It is a huge undertaking and the EPA staff has clearly worked hard. I continue to applaud the creation of a multi-pollutant, multi media standard that is linked to deposition and ecosystem responses.

### **General Comments:**

This version contains much clearer descriptions and general justification for the various aspects of the standard. And, while I think that a standard that is applied to 80+ ecoregions will present challenges, it is far more realistic given that ecosystem responses to atmospheric deposition are modulated by chemical, physical and biological features of the landscape, and that these features also vary across the landscape. That the coastal plain regions showed up prominently (Chapter 7, Figures in Appendix D) in the preliminary assessment was surprising (vs the Appalachians). While I agree with the discussion in Chapter 7 about the likely ecological reasons for this (organic vs mineral acids), I was less clear what the practical implications are, and how, specifically, the coastal plains regions might be dealt with in regard to the standard.

The uncertainty discussion is much improved in chapter 7. As expected there are considerable uncertainties in all aspects of the AAI—it is the state of the science and the state of the models. The goal of reducing uncertainties and further testing and refining both the models and data collection in the future (esp. section F-50) is quite important. I found interesting, and not too surprising that, in general, base cation weathering, and Q, for most locations, had a greater influence on AAI than transference ratios (Appendix, page F-11 in final version of PAD). These results highlight research and monitoring needs.

The AAI is an innovative and ecologically relevant form. It considers the complex interactions of ecological systems from emissions to deposition to ecological effects via complicated biogeochemical reactions within ecosystems. There is much modeled and measured information and data packed within the AAI, as we have discussed at length in the past, including consideration of NH<sub>x</sub> inputs to ecosystems.

Atmospheric NO<sub>y</sub> and SO<sub>x</sub> are the indicators that this standard has to “work with.” As noted throughout this review, other forms of N (e.g., NH<sub>x</sub>) are important to ecological function, on the rise, and should be considered explicitly (not just as an “offset” in the AAI) in the future. ANC is a scientifically defensible indicator of aquatic acidification, and its proposed (range) level is appropriately supported by the literature.

Multiyear averaging time to smooth out anomalous years makes sense. However, it does not make complete sense to me to run CMAQ on a daily time step when 3-5 year averaging would be used. Also, I don't completely buy the argument that it's the temporal, rather than the spatial heterogeneity that is most important to capture in modeling deposition, especially since CMAQ still cannot resolve finer (spatial) scale deposition to heterogeneous, ecologically sensitive mountainous regions. An important goal should be to generate annual deposition over finer spatial scales and complex terrain. (I note that there is vague reference to doing this in section F.5 of the Appendices—more detail would have been useful.)

I cannot stress enough the importance of gathering spatially explicit monitoring data and stimulating and supporting ecosystem based research to fill the significant gaps we have identified during this process, from inputs (deposition) to biogeochemical processes within and among ecosystems. I should hope that the next review will be made much more straightforward, and uncertainties will be much reduced as a result.