



# Policy Assessment for the Review of the Secondary National Ambient Air Quality Standards for NO<sub>x</sub> and SO<sub>x</sub>

Second External Review Draft

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**Policy Assessment for the Review of the  
Secondary National Ambient Air Quality Standards  
for NO<sub>x</sub> and SO<sub>x</sub>:**

**Second External Review Draft**

U.S. Environmental Protection Agency  
Office of Air Quality Planning and Standards  
Health and Environmental Impacts Division  
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## **DISCLAIMER**

This document has been reviewed by the Office of Air Quality Planning and Standards, U.S. Environmental Protection Agency (EPA), and approved for publication. This draft document has been prepared by staff from the Office of Air Quality Planning and Standards, U.S. Environmental Protection Agency. Any opinions, findings, conclusions, or recommendations are those of the authors and do not necessarily reflect the views of the EPA. Mention of trade names or commercial products is not intended to constitute endorsement or recommendation for use. This document is being provided to the Clean Air Scientific Advisory Committee for their review, and made available to the public for comment. Any questions or comments concerning this document should be addressed to Bryan Hubbell, U.S. Environmental Protection Agency, Office of Air Quality Planning and Standards, C504-06, Research Triangle Park, North Carolina 27711 (email: [hubbell.bryan@epa.gov](mailto:hubbell.bryan@epa.gov)).

**Acknowledgements (if any)**

# **Policy Assessment for the Review of the Secondary National Ambient Air Quality Standards for NO<sub>x</sub> and SO<sub>x</sub>: Second External Review Draft**

## **EXECUTIVE SUMMARY**

### **Introduction**

This second draft Policy Assessment is an evaluation of the policy implications of the key scientific information contained in the Integrated Science Assessment (ISA) for Oxides of Nitrogen and Sulfur-Ecological Criteria, prepared by EPA's National Center for Environmental Assessment (NCEA), and the results from the analyses contained in the Risk and Exposure Assessment (REA) for Review of the Secondary National Ambient Air Quality Standards for Oxides of Nitrogen and Oxides of Sulfur. This second draft also presents preliminary EPA staff conclusions regarding the adequacy of the current standards and various policy options that we believe are appropriate to consider as part of the current review of the secondary (welfare-based, e.g. focused on non-health effects including impacts on soils, water, crops, vegetation, man-made materials, animals, wildlife, weather, visibility, and climate) NO<sub>x</sub> and SO<sub>x</sub> NAAQS.

This policy assessment is intended to help "bridge the gap" between the scientific assessment contained in the ISA and the judgments required of the EPA Administrator in determining whether, and if so, how, it is appropriate to revise the secondary NAAQS for NO<sub>x</sub> and SO<sub>x</sub>. This policy assessment considers the available scientific evidence and quantitative risk-based analyses, together with related limitations and uncertainties, and focuses on the basic elements of air quality standards: indicators, averaging times, forms, and levels. These elements, which serve to define each standard, must be considered collectively in evaluating the welfare protection afforded by the secondary NO<sub>x</sub> and SO<sub>x</sub> NAAQS.

In conducting this periodic review of the NO<sub>x</sub> and SO<sub>x</sub> secondary NAAQS, EPA has decided to jointly assess the scientific information, associated risks, and standards because ambient NO<sub>x</sub> and SO<sub>x</sub>, and their associated transformation products, such as deposited N and S, are linked from an atmospheric chemistry perspective, as well as jointly contributing to environmental effects.

### **Scope**

This assessment primarily focuses on the effects of the deposition of ambient NO<sub>x</sub> and SO<sub>x</sub> on multiple ecological receptors. Highlighted effects include those associated with acidification and nitrogen nutrient enrichment. Based on these highlighted effects, EPA's objective is to develop a framework for setting standards that are ecologically relevant and that reflect the common impacts of these two pollutants as they deposit to sensitive ecosystems.

For this second draft policy assessment, we have chosen to focus much of our discussion on effects in sensitive aquatic ecosystems caused by acidifying deposition of nitrogen and sulfur, which is a transformation product of ambient NO<sub>x</sub> and SO<sub>x</sub>. We have a high degree of confidence in the link to aquatic acidification effects as well as more information available with which to develop an ecologically meaningful structure for the standards. We recognize in doing so that the resulting standards will not likely provide full protection against terrestrial acidification effects or against adverse nutrient enrichment effects in sensitive terrestrial and aquatic ecosystems. It is however likely that some additional protection for those ecosystems will be provided as overall NO<sub>x</sub> and SO<sub>x</sub> levels in the environment are decreased in response to the aquatic acidification based standards.

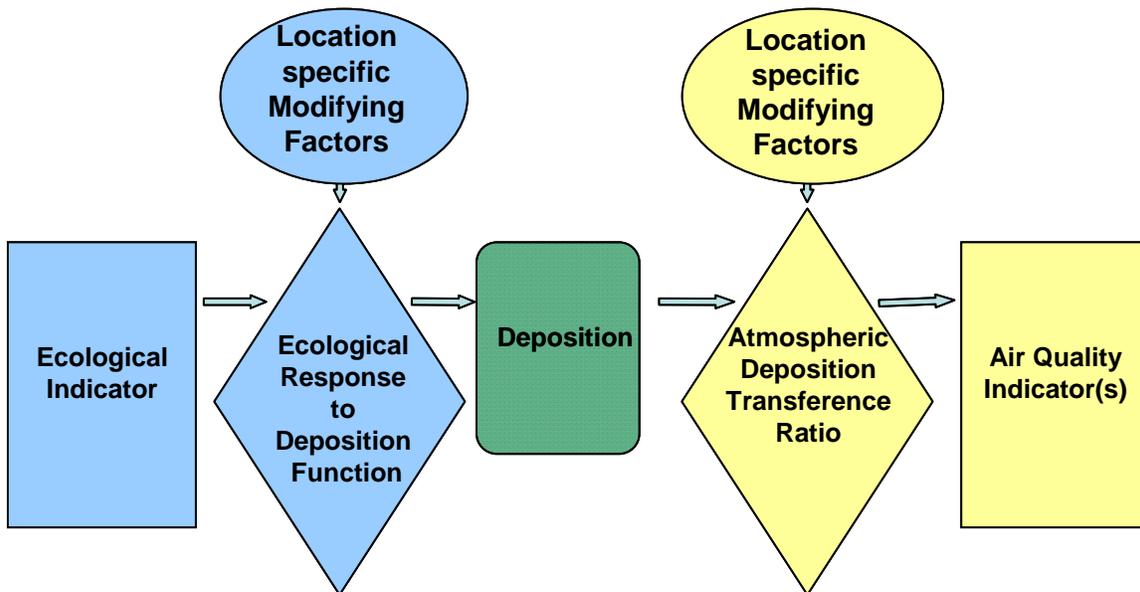
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In the atmospheric science community, NO<sub>x</sub> typically refers to the sum of nitrogen dioxide (NO<sub>2</sub>), and nitric oxide (NO). In contrast, the Clean Air Act uses “NO<sub>x</sub>” to refer to any gaseous mixture of species composed solely of nitrogen and oxygen (e.g., NO<sub>2</sub>, NO, nitrous oxide [N<sub>2</sub>O], nitrogen trioxide [N<sub>2</sub>O<sub>3</sub>], nitrogen tetroxide [N<sub>2</sub>O<sub>4</sub>], and dinitrogen pentoxide [N<sub>2</sub>O<sub>5</sub>]). The term used by the scientific community to represent the complete set of oxidized nitrogen compounds, including those listed in CAA Section 108(c), is total oxidized nitrogen (NO<sub>y</sub>). NO<sub>y</sub> includes all nitrogen oxides, including NO, NO<sub>2</sub>, HNO<sub>3</sub>, peroxyacetyl nitrate (PAN), 2N<sub>2</sub>O<sub>5</sub>, HONO, NO<sub>3</sub>, organic nitrates, and particulate NO<sub>3</sub>. In the policy assessment, unless otherwise indicated, we use the term “NO<sub>y</sub>” to refer to the complete set of oxidized nitrogen compounds.

For this assessment, the full definition of SO<sub>x</sub> includes all oxides of sulfur, including both gaseous substances (e.g., SO<sub>2</sub>, sulfur monoxide [SO], sulfur trioxide [SO<sub>3</sub>], thiosulfate [S<sub>2</sub>O<sub>3</sub>], and heptoxide [S<sub>2</sub>O<sub>7</sub>]), as well as particulate species, such as ammonium sulfate [(NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub>]). However, throughout this document we refer to SO<sub>x</sub> as the sum of SO<sub>2</sub> and sulfate to be consistent with standard monitoring instrumentation. Sulfate is referred to as SO<sub>4</sub> and nitrate as NO<sub>3</sub>, recognizing that they refer to the ions that have charges of -2 for sulfate and -1 for nitrate.

### Conceptual Framework

The figure below depicts the framework by which we are considering the structure of an ecologically relevant secondary standard. It is a conceptual diagram that illustrates how a level of protection related to an indicator of ecological effect(s) equates to atmospheric concentrations of NO<sub>x</sub> and SO<sub>x</sub> indicators. This conceptual diagram illustrates the linkages between ambient air concentrations and resulting deposition metrics, and between the deposition metric and the ecological indicator of concern. The Deposition Transference Ratios translate between NO<sub>x</sub> and SO<sub>x</sub> deposition metrics and ambient atmospheric concentrations of NO<sub>x</sub> and SO<sub>x</sub>, while the Ecological Response to Deposition Function relates the deposition metric into the ecological indicator.



Our policy assessment is structured around this conceptual model, and includes an evaluation of the effects associated with deposition of NO<sub>x</sub> and SO<sub>x</sub> to ecosystems, as well as an assessment of the

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adequacy of the existing NO<sub>x</sub> and SO<sub>x</sub> standards in protecting against these effects. This policy assessment also develops a more complete understanding of the conceptual structure needed to address the variable ecosystem and atmospheric factors which modify the impacts of deposited NO<sub>x</sub> and SO<sub>x</sub> on ecosystems. Development of the form for the standard and options for ambient atmospheric indicators for NO<sub>x</sub> and SO<sub>x</sub>, averaging times, and levels of the standard are also discussed.

### **Ecological Effects from NO<sub>x</sub> and SO<sub>x</sub> Deposition**

Effects are broadly categorized into those related to acidification and nutrient-enrichment. Acidification occurs in both aquatic and terrestrial ecosystems, with most aquatic effects occurring in freshwater lakes and streams. Nutrient enrichment also occurs in both aquatic and terrestrial ecosystems; however, the types and prevalence of nutrient enrichment effects vary between freshwater and estuarine aquatic ecosystems.

In the process of acidification, chemical components of terrestrial and freshwater aquatic ecosystems are altered in a way that leads to effects on biological organisms. Because NO<sub>x</sub> and SO<sub>x</sub> deposited to terrestrial ecosystems often move through the soil and eventually leach into adjacent water bodies, deposition to terrestrial ecosystems is also a cause of acidification in aquatic ecosystems.

The scientific evidence is sufficient to infer a strong causal relationship between acidifying deposition and effects on biogeochemical processes and biota in aquatic ecosystems, and between acidifying deposition and changes in biogeochemistry in terrestrial ecosystems. Acidic deposition is observed to alter sulfate and nitrate concentrations in surface waters, balance of base cations, acid neutralizing capacity (ANC), inorganic aluminum, calcium, and surface water pH. These changes can result in the loss of acid-sensitive biological species such as salmonid fish species and disrupt food web dynamics causing alteration to the diet, breeding distribution and reproduction of certain species of bird, such as goldeneye ducks and loons. Acidification in terrestrial ecosystems has been shown to cause decreased growth and increased susceptibility to disease and injury in sensitive tree species, including red spruce and sugar maple.

Principal factors governing the sensitivity of terrestrial and aquatic ecosystems to acidification from sulfur and nitrogen deposition include geology, biological uptake of nitrogen, soil depth, and elevation. Geologic formations having low base cation supply generally underlie the watersheds of acid-sensitive lakes and streams. Other factors that contribute to the sensitivity of soils and surface waters to acidifying deposition include topography, soil chemistry, land use, and hydrology. Episodic and chronic acidification tends to occur primarily (but not exclusively) at relatively high elevations in areas that have base-poor bedrock, high relief, and shallow soils.

Based on published analyses of surface water data from freshwater ecosystem surveys and monitoring, the most sensitive lakes and streams are located in New England, the Adirondack Mountains, the Appalachian Mountains (northern Appalachian Plateau and Ridge/Blue Ridge region), the Upper Midwest and high elevation Western ecosystems.

ANC is the most widely used indicator of acid sensitivity and has been found in various studies to be the best single indicator of the biological response and health of aquatic communities in acid-sensitive systems. Annual or multiyear average ANC is a good overall indicator of sensitivity, capturing the ability of an ecosystem to withstand episodic events such as spring melting that can lower ANC over shorter time spans. Biota are generally not harmed when annual average ANC levels are >100 microequivalents per liter (µeq/L). At annual average ANC levels between 100 and 50 µeq/L, the fitness of sensitive species (e.g., brook trout, zooplankton) begins to decline. When annual average ANC is <50 µeq/L,

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negative effects on aquatic biota are observed, including large reductions in diversity of fish species, and declines in health of fish populations, affecting reproductive ability and fitness. Annual average ANC levels below 50 µeq/L are generally associated with death or loss of fitness of biota that are sensitive to acidification.

Recent studies indicate that acidification of lakes and streams can result in significant loss in economic value, which is one indicator of adversity associated with loss of ecosystem services. A 2006 study of New York residents found that they are willing to pay between \$300 and \$800 million annually for the equivalent of improving lakes in the Adirondacks region to an ANC of 50. In addition, several states have set goals for improving the acid status of lakes and streams, generally targeting ANC in the range of 50 to 60 µeq/L, and have engaged in costly activities to decrease acidification.

Forests of the Adirondack Mountains of New York, Green Mountains of Vermont, White Mountains of New Hampshire, the Allegheny Plateau of Pennsylvania, and high-elevation forest ecosystems in the southern Appalachians are the regions most sensitive to terrestrial acidification effects from acidifying deposition. A commonly used indicator of terrestrial acidification is the base cation to aluminum ratio, Bc/Al. Many locations in sensitive areas of the U.S. have Bc/Al levels below benchmark levels we have classified as providing low to intermediate levels of protection to tree health. At a Bc/Al ratio of 1.2 (intermediate level of protection), red spruce growth can be reduced by 20 percent. At a Bc/Al ratio of 0.6 (low level of protection), sugar maple growth can be reduced by 20 percent. While not defining whether a 20 percent reduction in growth can be considered significant, existing economic studies suggest that avoiding significant declines in the health of spruce and sugar maple forests may be worth billions of dollars to residents of the Eastern U.S.

The numerous ecosystem types that occur across the U.S. have a broad range of sensitivity to N deposition. Organisms in their natural environment are commonly adapted to a specific regime of nutrient availability. Change in the availability of one important nutrient, such as N, may result in imbalance in ecosystems, with effects on ecosystem processes, structure and function. In certain N-limited ecosystems, including many ecosystems managed for commercial production, N deposition can result in beneficial increases in productivity. Nutrient enrichment effects from NO<sub>x</sub> deposition are difficult to disentangle from overall effects of nitrogen enrichment. This is caused by two factors: the inputs of reduced nitrogen from deposition and, in estuarine ecosystems, a large fraction of nitrogen inputs from non-atmospheric sources.

### **Adequacy of the Existing NO<sub>x</sub> and SO<sub>x</sub> Standards to Protect Against Acidification and Nutrient Enrichment Effects**

Current NO<sub>x</sub> and SO<sub>x</sub> secondary standards are designed to protect against direct exposure of vegetation to ambient concentrations of NO<sub>x</sub> and SO<sub>x</sub>. Almost all areas of the U.S. are in attainment of the current NO<sub>x</sub> and SO<sub>x</sub> secondary standards. The NO<sub>x</sub> secondary standard is 0.053 parts per million (ppm), annual arithmetic average, calculated as the arithmetic mean of the 1-hour NO<sub>2</sub> concentrations. The SO<sub>x</sub> secondary standard, which uses SO<sub>2</sub> as the atmospheric indicator, is a 3-hour average of 0.5 ppm, not to be exceeded more than once per year.

Recent acidification status of aquatic ecosystems indicate that in the Adirondacks and Shenandoah areas, rates of acidifying deposition of NO<sub>x</sub> and SO<sub>x</sub> are still well above pre-acidification (1860) conditions. Forty-four percent of Adirondack lakes evaluated exceed the critical load for an ANC of 50 µeq/L, and in these lakes recreationally important fish species such as trout are missing due to acidification. In the Shenandoah area, 85 percent of streams evaluated exceed the critical load for an ANC of 50 µeq/L, resulting in losses in fitness in species such as the Blacknose Dace.

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The REA only evaluated a small number of sensitive areas as case studies. However, in the sugar maple case study area (Kane Experimental Forest, Pennsylvania), recent (2002) deposition levels are associated with a Bc/Al ratio below 1.2, indicating the potential for a greater than 20 percent reduction in growth. In the red spruce case study area (Hubbard Brook Experimental Forest, New Hampshire), recent deposition levels are associated with a Bc/Al ratio slightly above 1.2, indicating slightly less potential for significant reductions in growth.

Available ecological indicators for estuarine nutrient enrichment are not sufficiently sensitive to changes in atmospheric NO<sub>x</sub> to be of use in assessing the adequacy of existing NO<sub>x</sub> standards. Atmospheric NO<sub>x</sub> can be an important contributor of N to estuarine nutrient enrichment, but additional analysis is required to develop an appropriate indicator for assessing levels of protection from nutrient enrichment effects in estuaries related to deposition of NO<sub>x</sub>.

Nitrogen deposition can alter species composition and cause eutrophication in freshwater systems. In the Rocky Mountains, for example, deposition loads of 1.5 to 2 kg/ha/yr, which are within the range associated with ambient NO<sub>x</sub> levels meeting the current standards, are known to cause changes in species composition in diatom communities indicating impaired water quality. From this we initially conclude that the existing secondary standard for NO<sub>x</sub> does not protect such ecosystems and their resulting services from impairment.

Most terrestrial ecosystems in the US are N-limited, and therefore they are sensitive to perturbation caused by N additions. Under recent conditions, nearly all of the known sensitive mixed conifer forest ecosystems receive total N deposition levels above 3.1 N kg/ha/yr, which is the ecological benchmark for changes in lichen species. Lichens are sentinels for broader ecosystem change in terrestrial systems. Some portions of the Sierra Nevadas receive total N deposition levels above 5.2 N kg/ha/yr, which is the ecological benchmark for shifts in the dominant species of lichen from acidophytic to tolerant species. In addition, in Coastal Scrub Sage ecosystems in California, N deposition exceeds the 3.3 N kg/ha/yr benchmark above which nitrogen is no longer a limiting nutrient, leading to potential alterations in ecosystem composition. Because excessive N deposition and effects are observed in areas where, under recent conditions, NO<sub>x</sub> ambient concentrations are at or below the current NO<sub>x</sub> secondary standards, we initially conclude those standards are not adequate to protect against anticipated adverse impacts from N nutrient enrichment in sensitive ecosystems (systems where N is limiting) that are not managed for commercial agricultural and forest production.

Sulfur deposition is also linked with the formation of methylmercury. The production of methylmercury in aquatic ecosystems requires sulfate as well as mercury. The evidence is sufficient to infer a causal relationship between sulfur deposition and increased mercury methylation in wetlands and aquatic environments. However, while the production of methylmercury requires the presence of sulfate and mercury, the amount of methylmercury produced varies with oxygen content, temperature, pH, and supply of labile organic carbon. Due to limits in data, we are unable to assess the adequacy of the existing standards in protecting against effects associated with increased mercury methylation.

### **Conceptual Design of an Ecologically Relevant Standard**

The overall concept for ecologically relevant standards recognizes that the fundamental welfare effects associated with ambient NO<sub>x</sub> and SO<sub>x</sub> occur through the process of deposition to sensitive ecosystems. There are four main components to the conceptual design of the standard: atmospheric and ecological indicators, deposition metrics, functions that relate indicators to deposition metrics and factors that modify the functions. In this policy assessment, the focus is on developing the conceptual design for a

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standard that protects against effects associated with acidifying deposition of NO<sub>x</sub> and SO<sub>x</sub> in aquatic ecosystems, but this general conceptual framework is intended to apply to a broader set of potential endpoints.

For the conceptual design of an aquatic acidification standard, ANC is suggested as the ecological indicator. ANC is suggested as the ecological indicator because it is the most widely used chemical indicator of acid sensitivity in aquatic ecosystems and has been found through numerous studies to be the best single indicator of the biological response and health of aquatic communities in acid-sensitive systems. Furthermore, ANC can be directly linked to both underlying water chemistry, e.g. pH and aluminum, and to biological impairment, specifically the number of fish species in a water body.

Acidification models represent the ecological response relationship between ANC and deposition of N and S. Acidification models are designed for the catchment scale. However, for consideration of a national standard, aggregation to a broader spatial scale is desirable; therefore a method to evaluate critical loads (the levels of deposition of N and S below which defined levels of harmful effects on specified sensitive elements of the environment do not occur) across the national landscape is presented. The atmospheric transformation functions then convert deposition to ambient concentrations of NO<sub>x</sub> and SO<sub>x</sub>.

Acidification models relate ANC to deposition of N and S at the catchment scale. We suggest using an acidification model that incorporates environmental variables that modify the ecological response relationship. This includes a variable to account for nitrogen uptake by ecosystems. The acidification model can be used to calculate critical loads for individual catchments based on a selected level of ANC. The load of deposition that causes a selected level of ANC varies across the nation depending on the characteristics of the catchment, such as base cation weathering rates, nitrogen retention and the level of naturally occurring organic acids.

Although critical loads for a selected level of ANC will vary catchment by catchment it is not practical for a national standard to evaluate every catchment in the U.S. Therefore, we propose two general approaches to establish critical loads. One approach is to develop a national distribution of critical loads over all levels of sensitivity, recognizing that there is a high degree of heterogeneity in acid sensitivity even at relatively small spatial scales. The second approach is to subdivide the landscape of the U.S. into acid-sensitivity categories, such that within a category there are generally similar acid sensitivity characteristics. Each national acid-sensitivity category is represented by a population of catchments for which critical loads at a specified ANC limit are calculated. This second draft policy assessment explores a number of methods for developing the acid sensitivity categories.

National acid-sensitivity categories should be based on features that govern ecological sensitivity to acidification. Areas that have similar underlying geology, mineral weathering rates, and hydrology should show similar sensitivity to NO<sub>x</sub> and SO<sub>x</sub> deposition. Ecoregions are useful geographic definitions that holistically incorporate a number of important factors related to acid sensitivity, including geology, physiography, vegetation, climate, soils, land use, and hydrology. As such, the determination of acid-sensitivity categories begins with aggregation of lakes and streams by ecoregion. From there, acid sensitivity is further characterized using measured ANC. As noted earlier, ANC is a good indicator of the overall sensitivity of a water body to acidification. ANC measurement is also widely available in the U.S., making it a useful metric for further classifying ecoregions. The policy assessment explores several methods for using ANC to classify ecoregions using both simple and more complex methods.

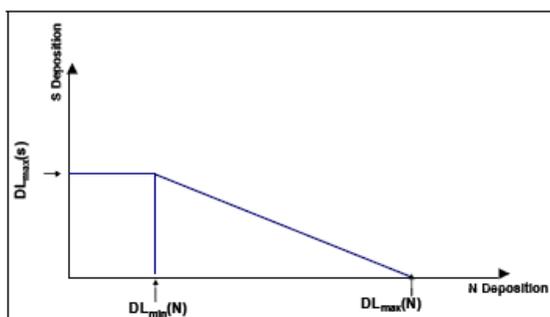
Once acid-sensitivity categories are defined, a sample of catchments will be selected to represent each category. The acidification models will be used to evaluate the critical load for each catchment in the

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population at the selected level of ANC. There is a distribution of critical load values within each acid-sensitivity category reflecting the range of sensitivity of catchments within each category. The goal of aggregating critical loads from multiple catchments is to develop an appropriately representative deposition value based on the distribution of critical loads, called the deposition metric, which protects a percentage of the population of water bodies within a national acid-sensitivity category from exceeding their critical load for the target value of ANC.

Distributions of catchment level critical loads are based on a combination of previously conducted steady-state critical load modeling and new critical load modeling conducted as part of this policy assessment. To ensure the population of water bodies included in the analysis were those sensitive to acidity caused by atmospheric deposition, several criteria were applied to the critical loads dataset to remove catchments in which organic acids, acid mine drainage or naturally low base cation weathering caused acidification.

Once a deposition metric is calculated, the value is modified by addition of a term to represent the amount of N that will be taken up by vegetation, immobilized in soil or degassed from the ecosystem. Next, a tradeoff curve for N and S deposition is generated for the acid sensitivity category. This function (illustrated below) is characterized by three nodes: 1) the maximum of amount of N deposition when S deposition equals zero 2) the amount of N deposition that will be captured by the ecosystem before it leaches and 3) the maximum amount of S sulfur deposition considering the N captured by the ecosystem. The function represents all pairs of N and S deposition that will equal the deposition metric for acidifying deposition for a specific target ANC.



**The depositional load function**

Reduced forms of nitrogen, such as ammonia, are either taken up by plants and microbes or converted to nitrate in the environment and use up the assimilative capacity of ANC at the same rate as oxidized forms of nitrogen deposition; therefore, deposition of reduced nitrogen must be accounted for in the watershed. The suggested approach is to subtract the loadings of reduced forms of nitrogen derived for a given spatial area from the deposition metric that represents a selected percentage of critical loads for a given population, such that the resultant deposition metric is for sulfur and oxidized nitrogen only. The policy assessment explores several methods for specifying the loadings of reduced nitrogen for specific geographic areas.

Deposition is related to ambient concentrations of NO<sub>x</sub> and SO<sub>x</sub> through deposition velocity, which is the rate at which an ambient pollutant is deposited. Deposition velocity varies over time and location, and is affected by land use conditions and meteorology. Our conceptual model requires conversion of deposition of N and S into ambient concentrations of NO<sub>x</sub> and SO<sub>x</sub>. Since the policy objective is to set an ambient air quality standard for total oxidized sulfur and nitrogen, and this is also the chemical resolution provided by the ecosystem models, it is convenient to use conversion factors based on available estimates

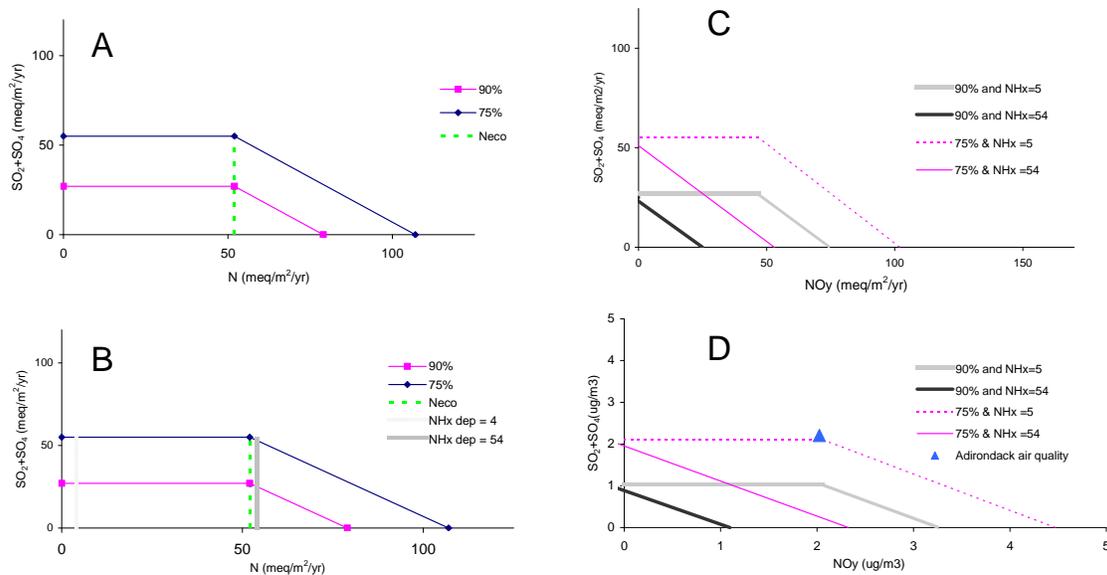
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of concurrent deposition and ambient concentrations. The ratio of total deposition of NO<sub>x</sub> and SO<sub>x</sub> to ambient concentrations of NO<sub>x</sub> and SO<sub>x</sub>, respectively are referred to as deposition transference ratios. Applying the estimated deposition transference ratios to depositional load tradeoff curves leads to parallel tradeoff curves for ambient NO<sub>x</sub> and SO<sub>x</sub> (see figure below).

The national ambient air quality standard must be able to tie ANC to deposition and deposition to ambient air concentrations, incorporating ecological conditions and the contribution of reduced nitrogen. To incorporate all of these aspects, we develop an index that will provide a consistent standard nationally that is directly expressed in terms of concentrations of NO<sub>x</sub> and SO<sub>x</sub>. The form of this standard is referred to as the Atmospheric Acidification Protection Index (AAPI), which can be applied across the nation to convey the protection of aquatic ecosystems from acidification due to atmospheric deposition.

The AAPI represents the level of protection against the effects of acidification given local ecological conditions, the level of reduced nitrogen being deposited, and the levels of NO<sub>x</sub> and SO<sub>x</sub> that are limited by the standard. The AAPI is linked to a target ANC for a chosen percent of lakes and streams, and determines the combinations of NO<sub>x</sub> and SO<sub>x</sub> that will jointly result in the target ANC, taking into account uncertainties and other factors.

This AAPI can also be expressed as the tradeoff curves which show the combinations of NO<sub>x</sub> and SO<sub>x</sub> that meet the standard, generated for specific values of AAPI, and provide a representation of the standards in terms of atmospheric concentrations. These tradeoff curves will vary across the U.S. based on ecosystem sensitivity, reduced nitrogen deposition levels, and other factors. An example tradeoff curve for two different percentiles of protection at a target ANC of 50 µeq/L is provided below.



Graphs demonstrate the steps to develop a tradeoff curve for the AAPI including A) N vs. SO<sub>2</sub>+SO<sub>4</sub> from the deposition metric and modified with Neco B) Two loads of NHx indicated C) after subtracting NHx load the resultant NOy vs SO<sub>2</sub>+SO<sub>4</sub> curve, D) After applying the transference ratio the resultant air concentrations.

## Options for Elements of the Standards

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Ambient air quality indicators other than NO<sub>2</sub> and SO<sub>2</sub> should be considered as the appropriate pollutant indicators for protection against the acidification effects associated with deposition of NO<sub>x</sub> and SO<sub>x</sub>. This consideration is based on the recognition that all forms of oxidized nitrogen and sulfur in the atmosphere contribute to deposition and resulting acidification, and as such concentrations of NO<sub>2</sub> and SO<sub>2</sub> are incomplete indicators. Furthermore, concentration of NO<sub>y</sub> is proposed as an appropriate indicator for oxides of nitrogen. The sum of concentrations of SO<sub>2</sub> and SO<sub>4</sub> is proposed as an appropriate indicator for oxides of sulfur.

Welfare effects associated with acidification result from annual cumulative deposition of nitrogen and sulfur, reflected in effects on the chronic ANC level (measured as annual or multiyear average ANC). Short-term (i.e., hours or days) episodic changes in water chemistry can also have significant biological effects. Episodic chemistry refers to conditions during precipitation or snowmelt events. Surface water chemistry has lower pH and acid neutralizing capacity (ANC) during these events than under baseflow conditions. One of the most important effects of acidifying deposition on surface water chemistry is the short-term change in chemistry that is termed “episodic acidification.” While ecosystems are also affected by episodic increases in acidity due to pulses of acidity during high rainfall periods and snowmelts, protection against these episodic acidity events can be achieved by establishing a higher chronic ANC level. Episodic acidification can result from either shorter term deposition episodes, or from longer term deposition on snowpack. Snowmelt can release stored N deposited throughout the winter, leading to episodic acidification in the absence of increased deposition during the actual episodic acidification event. Long term (3 to 5 year average) ambient NO<sub>x</sub> and SO<sub>x</sub> concentrations are appropriate to provide protection against low chronic ANC levels and episodic acidification.

The current forms of the secondary standards for NO<sub>x</sub> and SO<sub>x</sub> do not take into account the combined contributions of NO<sub>x</sub> and SO<sub>x</sub> in the causation of effects associated with acidification of aquatic ecosystems. Based on the causal linkages between NO<sub>x</sub> and SO<sub>x</sub>, deposition of N and S, and the indicator of acidification, ANC, the current forms should be replaced with an atmospheric acidification protection index (AAPI), which reflects the important roles of underlying ecosystem characteristics, determinants of deposition, and deposition of reduced nitrogen in determining the potential effects from deposition of NO<sub>x</sub> and SO<sub>x</sub>.

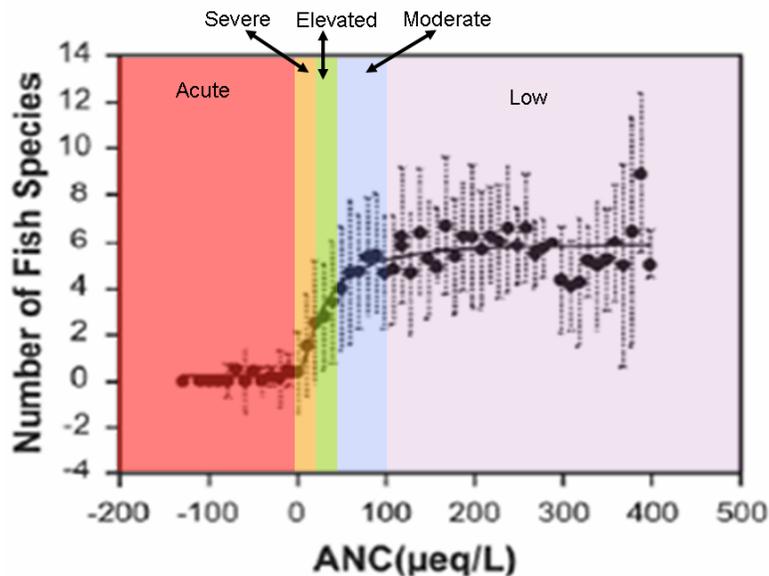
The value of AAPI can be calculated for any observed values of NO<sub>x</sub> and SO<sub>x</sub>. However, the level of the standard for AAPI should reflect a wide number of factors, including desired level of protection indicated by a target ANC limit, the target percentile of water bodies to achieve the target ANC, and the various factors and uncertainties involved in specifying all of the other aspects of the standard, such as the acid sensitivity classification method, the specification of deposition of reduced nitrogen, nitrogen retention, the deposition transference ratios, and the averaging time. The administrator may choose an AAPI level reflecting an ANC level higher or lower than the target ANC limit to account for the combined effect of all of the components of the standard and their related uncertainties. The resulting AAPI, in the context of the overall standard, will reflect her informed judgment as to a standard that is sufficient but not more than necessary to protect against adverse public welfare effects.

Within the AAPI form, EPA will specify the parameter values for all elements excepting NO<sub>x</sub> and SO<sub>x</sub>, which will be the measured atmospheric indicators. The values for pre-industrial base cation weathering, nitrogen retention and uptake, and runoff, are based on the same inputs used to develop the deposition metrics. The values for reduced nitrogen and the deposition transference ratios will be calculated using output from the most up-to-date version of EPA’s Community Multiscale Air Quality (CMAQ) model. EPA is considering methods to account for the dynamic nature of deposition of reduced nitrogen in specifying the NO<sub>x</sub> and SO<sub>x</sub> standards.

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As demonstrated by the tradeoff curves, multiple combinations of concentrations of NO<sub>x</sub> and SO<sub>x</sub> can yield the same value of the AAPI. No single combination of NO<sub>x</sub> and SO<sub>x</sub> will produce a particular value of AAPI in all locations. Measured concentrations of annual average NO<sub>x</sub> and SO<sub>x</sub> necessary to meet the standards are thus expressed conditionally by the AAPI and not by fixed quantities.

A target ANC limit based on a desired level of protection is an important input to the decisions of the level of AAPI and the percent of ecosystems to be protected. Specific levels of ANC are associated with differing levels of ecosystem impairment, with higher levels of ANC resulting in fewer ecosystem impacts, and lower levels resulting in both higher intensity of impacts and a broader set of impacts. For example, the number of fish species present in a waterbody has been shown to be positively correlated with the ANC level in the water, with higher values supporting a greater richness and diversity of fish species. This relationship is illustrated in the following figure.



**Number of fish species per lake or stream versus ANC level and aquatic status category for lakes in the Adirondack Case Study Area**

The target ANC level specified in designing the standard is only one part in determining the overall protectiveness of the standard. The degree of protectiveness is based on all elements of the standard, including the target ANC selected by the Administrator, the size of the spatial areas over which the standard is applied, the percent of aquatic ecosystems targeted within a spatial area that is selected by the Administrator to achieve the selected ANC level, and the underlying parameters of the AAPI, including the atmospheric indicator, the critical load models used to determine pre-industrial base cation levels, the calculated values for the deposition transference ratios, and the calculated value for deposition of reduced nitrogen. There are widely varying degrees of uncertainty associated with all of these elements, some being much more certain and others being much less certain. The specified target ANC level is a crucial part of developing a standard that is requisite to protect public welfare, but it is the overall design and content of the standard that must be considered in judging the adequacy of protection it provides.

The secondary NAAQS will reflect the public welfare policy judgments of the Administrator, based on the science, as to the level of air quality which is requisite to protect the public welfare from any known or anticipated adverse effects associated with the pollutant in the ambient air. In certain naturally acidic ecosystems, even though the ecological benchmarks are exceeded, e.g. ANC may be quite low; NO<sub>x</sub> and

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SO<sub>x</sub> are not contributing to effects because those systems have chronic natural acidity and will not benefit from reductions in atmospheric deposition. The secondary NAAQS are not intended to provide protection in these types of naturally acidic systems. Instead, the secondary NAAQS are focused on providing protection in areas where ambient NO<sub>x</sub> and SO<sub>x</sub> are resulting in effects in ecosystems with low natural levels of acidification that are highly sensitive to additional inputs of acid deposition. The approaches for specifying populations of critical loads to develop deposition metrics explicitly excludes lakes and streams that are naturally acidic and those not likely to benefit from decreases in atmospheric deposition.

ANC levels below 20 µeq/L are generally associated with high probability of low pH, leading to death or loss of fitness of biota that are sensitive to acidification. Overall, there is little uncertainty that significant effects on aquatic biota are occurring at ANC levels below 20 µeq/L. Based on the field data from the Adirondacks and Shenandoah case study areas, ANC levels less than 50 µeq/L are adverse to ecosystem health, and are likely to lead to reductions in ecosystem services related to recreational fishing. However, the types of effects, specific species, and prevalence of effects across water bodies in the U.S. is more uncertain at ANC levels between 20 and 50 µeq/L. Targeting ANC levels between 50 and 100 µeq/L would provide additional protection; however, uncertainties regarding the additional reduction in adverse welfare effects are much larger for target ANC levels above 50 µeq/L.

Specifying an appropriate range of levels for an AAPI standard that is designed and specified as discussed above involves consideration of the degree to which any specific AAPI would lead to achieving the desired ANC level, and a judgment as to the degree of protection of public welfare that is warranted. Selection of a range of AAPI and selection of a specific level of AAPI within that range should incorporate a wide number of considerations, including the percent of water bodies within acid sensitive areas that the Administrator determines should be protected at the targeted ANC level.

In determining the requisite level of protection for the public welfare from effects on aquatic ecosystems, the Administrator will need to weigh the importance of the predicted risks of these effects in the overall context of public welfare protection, along with a determination as to the appropriate weight to place on the associated uncertainties and limitations of this information.

### **Co-Protection for Other Effects Provided by an Acidification Standard**

To understand the level of protection provided by a NO<sub>x</sub>/SO<sub>x</sub> standard based on aquatic acidification to protect against terrestrial acidification effects, we compared the critical loads for lakes and streams that would maintain an aquatic ANC of 50 to the critical loads to maintain either a terrestrial Bc:Al ratio of 1.2 or 10 averaged across a watershed area.

Results for the Adirondacks showed that critical loads for 29 lakes at an ANC of 50 were lower for 13 of those lakes than the critical load for the terrestrial watershed areas at a Bc:Al ratio of 10 and for 21 lakes at a Bc:Al ratio of 1.2. Perhaps more significant was the result that 13 of the 16 lakes in the highly and moderately sensitive areas had a lower critical load than the Bc:Al 10 areas and 16 of 16 lakes in the highly and moderately sensitive areas had lower critical loads than the Bc:Al 1.2 areas. The Shenandoah region reflected similar results.

In general, the aquatic critical acid loads offered greater protection to the watersheds than did the terrestrial critical loads. Generally in situations where the terrestrial critical loads were more protective, the lakes or streams in the watershed were rated as having “Low Sensitivity” or “Not Sensitive” to acidifying nitrogen and sulfur deposition. Conversely, when the water bodies were more sensitive to

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deposition (“Highly Sensitive” or “Moderately Sensitive”), the aquatic critical acid loads generally provided a greater level of protection against acidifying nitrogen and sulfur deposition in the watershed.

### **Initial Conclusions**

In this current review, important new information has become available since the last reviews (1996 for NO<sub>x</sub>, and 1988 for SO<sub>x</sub>) that supports revising the current NO<sub>x</sub> and SO<sub>x</sub> standards. Specifically, the ISA has concluded that there are causal relationships between NO<sub>x</sub> and SO<sub>x</sub> acidifying deposition and effects on aquatic and terrestrial ecosystems, and the ISA and REA provide substantial quantitative evidence of effects occurring in locations that meet the current NO<sub>2</sub> and SO<sub>2</sub> standards. In addition, observational data and rigorous atmospheric modeling has become available regarding the role of both nitrogen and sulfur deposition in acidification of sensitive water bodies. This information is sufficient to inform the development of revised secondary standards for NO<sub>x</sub> and SO<sub>x</sub> to protect against the effects of aquatic acidification in sensitive ecosystems. Additional information is needed to set separate standards to protect terrestrial ecosystems from acidification effects. While there is also new information available on the role of nitrogen deposition on nutrient enrichment effects in sensitive terrestrial and aquatic ecosystems, and the ISA concludes there is a causal relationship between NO<sub>x</sub> and nutrient enrichment effects, for this second draft policy assessment, we have focused on acidification effects due to the substantially greater amount of information available to inform the development of secondary standards.

We highlight the progress made in considering the joint nature of ecosystem responses to acidifying deposition of NO<sub>x</sub> and SO<sub>x</sub>, and note that the ability to consider revisions to the NO<sub>x</sub> and SO<sub>x</sub> secondary standards has been enhanced by our ability to consider a joint standard for NO<sub>x</sub> and SO<sub>x</sub> to protect against acidification effects. The development of an appropriate form of the standard linked to a common indicator of aquatic acidification, ANC, is also a significant step forward, as it allows for development of a standard for aquatic acidification designed to provide generally the same degree of protection across the country, while still reflecting the underlying variability in ecosystem sensitivity to acidifying NO<sub>x</sub> and SO<sub>x</sub> deposition.

We provide the following initial conclusions regarding the NO<sub>x</sub> and SO<sub>x</sub> secondary standards:

- The available effects-based evidence for aquatic and terrestrial acidification and nutrient enrichment suggests consideration of NO<sub>x</sub> and SO<sub>x</sub> standards that are at least as protective as the current standard. Consideration of joint standards for NO<sub>x</sub> and SO<sub>x</sub> is appropriate given the common atmospheric processes governing the deposition of NO<sub>x</sub> and SO<sub>x</sub> to sensitive ecosystems.
- On the basis of the acidification and nutrient enrichment effects that have been observed to still occur under current ambient conditions and those predicted to occur under the scenario of just meeting the current secondary NAAQS, we find support for consideration that the current secondary NAAQS are inadequate to protect the public welfare from known and anticipated adverse welfare effects from aquatic and terrestrial acidification associated with deposition of NO<sub>x</sub> and SO<sub>x</sub>.
- We find support for consideration that current levels of NO<sub>x</sub> and SO<sub>x</sub> are associated with deposition that leads to ANC values below benchmark values that cause ecological harm and losses in ecosystem services in sensitive ecosystems, including significant mortality in sensitive aquatic biota and losses in fish species richness, which is associated with reductions in recreational fishing services, among others.
- We find support for consideration that current levels of ambient NO<sub>x</sub> and SO<sub>x</sub> are associated with deposition that leads to Bc:Al values below benchmark values for terrestrial acidification that cause ecological harm and losses in ecosystem services in sensitive ecosystems, including losses

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in tree health and growth, which are associated with reductions in timber production, among other services.

- We suggest that effects due to aquatic acidification are most suitable for defining secondary standards for NO<sub>x</sub> and SO<sub>x</sub>. We note that in developing a standard designed to protect against the effects of aquatic acidification due to deposition of NO<sub>x</sub> and SO<sub>x</sub>, the resulting standards may not provide adequate protection against known effects associated with nutrient enrichment in sensitive aquatic and terrestrial ecosystems or acidification in sensitive terrestrial ecosystems.
- Based on the causal linkages between NO<sub>x</sub> and SO<sub>x</sub>, deposition of N and S, and the indicator of acidification, ANC, consideration should be given to an additional secondary standard with a form defined by an atmospheric acidification protection index (AAPi), which reflects the important roles of underlying ecosystem characteristics, determinants of deposition, and deposition of reduced nitrogen in determining the potential effects from deposition of NO<sub>x</sub> and SO<sub>x</sub>.
- Staff has concluded, based on the evidence and risk based information, and consideration of information related to definitions of adversity, that:
  - a target level of ANC of 20 µeq/L will protect against significant losses in fish mortality in many sensitive lakes, but will place less weight on protection against losses in aquatic biodiversity, and will be less protective against potential acidification episodes,
  - a target level of ANC of 50 µeq/L will protect against significant mortality in aquatic organisms and loss of fish health and biodiversity in sensitive lakes and streams, and will give weight to considerations of uncertainties in the time to recovery of aquatic ecosystems,
  - target levels of ANC above 50 µeq/L may provide additional protection against declines in fitness of sensitive species (e.g., brook trout, zooplankton), however, overall health of aquatic communities may not be impacted.

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## List of Acronyms and Abbreviations

AAPI	Atmospheric Acidification Potential Index
ADR	Adirondack Mountains of New York
Al <sup>3+</sup>	aluminum
ANC	acid neutralizing capacity
AQCD	Air Quality Criteria Document
AQRV	air quality related values
ASSETS EI	Assessment of Estuarine Trophic Status eutrophication index
Bc/Al	Base cation to aluminum ratio, also Bc:Al
C	carbon
Ca/Al	calcium to aluminum ratio
Ca <sup>2+</sup>	calcium
CAA	Clean Air Act
CASAC	Clean Air Scientific Advisory Committee
CASTNet	Clean Air Status and Trends Network
CCS	coastal sage scrub
Chl <i>a</i>	chlorophyll <i>a</i>
CLE	critical load exceedance
CMAQ	Community Multiscale Air Quality model
CSS	coastal sage scrub
CWA	Clean Water Act
DIN	dissolved inorganic nitrogen
DO	dissolved oxygen
DOI	U.S. Department of Interior
EMAP	Environmental Monitoring and Assessment Program
EPA	U.S. Environmental Protection Agency
FHWAR	fishing, hunting and wildlife associated recreation survey
FIA	Forest Inventory and Analysis National Program
FWS	Fish and Wildlife Service
GIS	geographic information systems
GPP	gross primary productivity
H <sup>+</sup>	hydrogen ion
H <sub>2</sub> O	water vapor
H <sub>2</sub> SO <sub>4</sub>	sulfuric acid
ha	hectare
HAB	harmful algal bloom
HFC	hydrofluorocarbon
Hg <sup>+2</sup>	reactive mercury
Hg <sup>0</sup>	elemental mercury
HNO <sub>3</sub>	nitric acid
HONO	nitrous acid
HUC	hydrologic unit code
IMPROVE	Interagency Monitoring of Protected Visual Environments
ISA	Integrated Science Assessment
K <sup>+</sup>	potassium

kg/ha/yr	kilograms per hectare per year
km	kilometer
LRMP	Land and Resource Management Plan
LTER	Long Term Ecological Monitoring and Research
LTM	Long-Term Monitoring
MAGIC	Model of Acidification of Groundwater in Catchments
MCF	Mixed Conifer Forest
MEA	Millennium Ecosystem Assessment
Mg <sup>2+</sup>	magnesium
N	nitrogen
N <sub>2</sub>	gaseous nitrogen
N <sub>2</sub> O	nitrous oxide
N <sub>2</sub> O <sub>3</sub>	nitrogen trioxide
N <sub>2</sub> O <sub>4</sub>	nitrogen tetroxide
N <sub>2</sub> O <sub>5</sub>	dinitrogen pentoxide
Na <sup>+</sup>	sodium
NAAQS	National Ambient Air Quality Standards
NADP	National Atmospheric Deposition Program
NAPAP	National Acid Precipitation Assessment Program
NAWQA	National Water Quality Assessment
NEEA	National Estuarine Eutrophication Assessment
NEP	net ecosystem productivity
NH <sub>3</sub>	ammonia gas
NH <sub>4</sub> <sup>+</sup>	ammonium ion
(NH <sub>4</sub> ) <sub>2</sub> SO <sub>4</sub>	ammonium sulfate
NH <sub>x</sub>	category label for NH <sub>3</sub> plus NH <sub>4</sub> <sup>+</sup>
NO	nitric oxide
NO <sub>2</sub>	nitrogen dioxide
NO <sub>2</sub> <sup>-</sup>	reduced nitrite
NO <sub>3</sub> <sup>-</sup>	reduced nitrate
NOAA	National Oceanic and Atmospheric Administration
NO <sub>x</sub>	nitrogen oxides
NO <sub>y</sub>	total oxidized nitrogen
NPP	net primary productivity
NPS	National Park Service
NRC	National Research Council
NSWS	National Surface Water Survey
NTN	National Trends Network
NTR	organic nitrate
O <sub>3</sub>	ozone
OAQPS	Office of Air Quality Planning and Standards
OW	Office of Water
PAN	peroxyacyl nitrates
PFC	perfluorocarbons
pH	relative acidity
ppb	parts per billion

ppm	parts per million
ppt	parts per trillion
PSD	prevention of significant deterioration
REA	Risk and Exposure Assessment
REMAP	Regional Environmental Monitoring and Assessment Program
S	sulfur
S <sub>2</sub> O <sub>3</sub>	thiosulfate
S <sub>2</sub> O <sub>7</sub>	heptoxide
SAV	submerged aquatic vegetation
SF <sub>6</sub>	sulfur hexafluoride
SMP	Simple Mass Balance
SO	sulfur monoxide
SO <sub>2</sub>	sulfur dioxide
SO <sub>3</sub>	sulfur trioxide
SO <sub>3</sub> <sup>2-</sup>	sulfite
SO <sub>4</sub>	wet sulfate
SO <sub>4</sub> <sup>2-</sup>	sulfate ion
SOM	soil organic matter
SO <sub>x</sub>	sulfur oxides
SPARROW	SPATIally Referenced Regressions on Watershed Attributes
SRB	sulfate-reducing bacteria
STORET	STORage and RETrieval
TIME	Temporally Integrated Monitoring of Ecosystems
TMDL	total maximum daily load
TP	total phosphorus
USFS	U.S. Forest Service
USGS	U.S. Geological Survey
µeq/L	microequivalents per liter
µg/m <sup>3</sup>	micrograms per cubic meter

## List of Key Terms

- Acidification:** The process of increasing the acidity of a system (e.g., lake, stream, forest soil). Atmospheric deposition of acidic or acidifying compounds can acidify lakes, streams, and forest soils.
- Air Quality Indicator:** The substance or set of substances (e.g., PM<sub>2.5</sub>, NO<sub>2</sub>, SO<sub>2</sub>) occurring in the ambient air for which the National Ambient Air Quality Standards set a standard level and monitoring occurs.
- Alpine:** The biogeographic zone made up of slopes above the tree line, characterized by the presence of rosette-forming herbaceous plants and low, shrubby, slow-growing woody plants.
- Acid Neutralizing Capacity:** A key indicator of the ability of water to neutralize the acid or acidifying inputs it receives. This ability depends largely on associated biogeophysical characteristics, such as underlying geology, base cation concentrations, and weathering rates.
- Arid Region:** A land region of low rainfall, where “low” is widely accepted to be less than 250 mm precipitation per year.
- Base Cation Saturation:** The degree to which soil cation exchange sites are occupied with base cations (e.g., Ca<sup>2+</sup>, Mg<sup>2+</sup>, K<sup>+</sup>) as opposed to Al<sup>3+</sup> and H<sup>+</sup>. Base cation saturation is a measure of soil acidification, with lower values being more acidic. There is a threshold whereby soils with base saturations less than 20% (especially between 10%–20%) are extremely sensitive to change.
- Ecologically Relevant Indicator:** A physical, chemical, or biological entity/feature that demonstrates a consistent degree of response to a given level of stressor exposure and that is easily measured/quantified to make it a useful predictor of ecological risk.
- Critical Load:** A quantitative estimate of an exposure to one or more pollutants, below which significant (as defined by the analyst or decision maker) harmful effects on specified sensitive elements of the environment do not occur, according to present knowledge.
- Denitrification:** The anaerobic reduction of oxidized nitrogen (e.g., nitrate or nitrite) to gaseous nitrogen (e.g., N<sub>2</sub>O or N<sub>2</sub>) by denitrifying bacteria.
- Dry Deposition:** The removal of gases and particles from the atmosphere to surfaces in the absence of precipitation (e.g., rain, snow) or occult deposition (e.g., fog).
- Ecological Risk:** The likelihood that adverse ecological effects may occur or are occurring as a result of exposure to one or more stressors (U.S. EPA, 1992).
- Ecological Risk Assessment:** A process that evaluates the likelihood that adverse ecological effects may occur or are occurring as a result of exposure to one or more stressors (U.S. EPA, 1992).
- Ecosystem:** The interactive system formed from all living organisms and their abiotic (i.e., physical and chemical) environment within a given area. Ecosystems cover a hierarchy of spatial scales and can comprise the entire globe, biomes at the continental scale, or small, well-circumscribed systems such as a small pond.
- Ecosystem Benefit:** The value, expressed qualitatively, quantitatively, and/or in economic terms, where possible, associated with changes in ecosystem services that result either directly or indirectly in improved human health and/or welfare. Examples of ecosystem benefits that derive from improved air quality include improvements in habitats for sport fish species, the quality of drinking water and recreational areas, and visibility.

**Ecosystem Function:** The processes and interactions that operate within an ecosystem.

**Ecosystem Services:** The ecological processes or functions having monetary or non-monetary value to individuals or society at large. These are (1) supporting services, such as productivity or biodiversity maintenance; (2) provisioning services, such as food, fiber, or fish; (3) regulating services, such as climate regulation or carbon sequestration; and (4) cultural services, such as tourism or spiritual and aesthetic appreciation.

**Eutrophication:** The process by which nitrogen additions stimulate the growth of autotrophic biota, usually resulting in the depletion of dissolved oxygen.

**Nitrogen Enrichment:** The process by which a terrestrial system becomes enhanced by nutrient additions to a degree that stimulates the growth of plant or other terrestrial biota, usually resulting in an increase in productivity.

**Nitrogen Saturation:** The point at which nitrogen inputs from atmospheric deposition and other sources exceed the biological requirements of the ecosystem; a level beyond nitrogen enrichment.

**Occult Deposition:** The removal of gases and particles from the atmosphere to surfaces by fog or mist.

**Semi-arid Regions:** Regions of moderately low rainfall, which are not highly productive and are usually classified as rangelands. “Moderately low” is widely accepted as between 100- and 250-mm precipitation per year.

**Sensitivity:** The degree to which a system is affected, either adversely or beneficially, by NO<sub>x</sub> and/or SO<sub>x</sub> pollution (e.g., acidification, nutrient enrichment). The effect may be direct (e.g., a change in growth in response to a change in the mean, range, or variability of nitrogen deposition) or indirect (e.g., changes in growth due to the direct effect of nitrogen consequently altering competitive dynamics between species and decreased biodiversity).

**Total Reactive Nitrogen:** This includes all biologically, chemically, and radiatively active nitrogen compounds in the atmosphere and biosphere, such as NH<sub>3</sub>, NH<sub>4</sub><sup>+</sup>, NO, NO<sub>2</sub>, HNO<sub>3</sub>, N<sub>2</sub>O, NO<sub>3</sub><sup>-</sup>, and organic compounds (e.g., urea, amines, nucleic acids).

**Valuation:** The economic or non-economic process of determining either the value of maintaining a given ecosystem type, state, or condition, or the value of a change in an ecosystem, its components, or the services it provides.

**Variable Factors:** Influences which by themselves or in combination with other factors may alter the effects on public welfare of an air pollutant (section 108 (a)(2))

(a) Atmospheric Factors: Atmospheric conditions that may influence transformation, conversion, transport, and deposition, and thereby, the effects of an air pollutant on public welfare, such as precipitation, relative humidity, oxidation state, and co-pollutants present in the atmosphere.

(b) Ecological Factors: Ecological conditions that may influence the effects of an air pollutant on public welfare once it is introduced into an ecosystem, such as soil base saturation, soil thickness, runoff rate, land use conditions, bedrock geology, and weathering rates.

**Vulnerability:** The degree to which a system is susceptible to, and unable to cope with, the adverse effects of NO<sub>x</sub> and/or SO<sub>x</sub> air pollution.

**Welfare Effects:** The effects on soils, water, crops, vegetation, man-made materials, animals, wildlife, weather, visibility, and climate; as well as damage to and deterioration of

property, hazards to transportation, and the effects on economic values and on personal comfort and well-being, whether caused by transformation, conversion, or combination with other air pollutants (Clean Air Act Section 302[h]).

**Wet Deposition:** The removal of gases and particles from the atmosphere to surfaces by rain or other precipitation.

# 1 INTRODUCTION

2  
3 The U.S. Environmental Protection Agency (EPA) is presently conducting a review of  
4 the secondary National Ambient Air Quality Standards (NAAQS) for oxides of nitrogen (NO<sub>x</sub>)  
5 and oxides of sulfur (SO<sub>x</sub>). The EPA's overall plan and schedule for this review were presented  
6 in the Integrated Review Plan for the Secondary National Ambient Air Quality Standards for  
7 Nitrogen Dioxide and Sulfur Dioxide (US EPA, 2007). The Integrated Review Plan (IRP)  
8 outlined the Clean Air Act (CAA or the Act) requirements related to the establishment and  
9 reviews of the NAAQS, the process and schedule for conducting the current review, and the key  
10 components in the NAAQS review process: an Integrated Science Assessment (ISA), Risk and  
11 Exposure Assessment (REA), and policy assessment/rulemaking. It presented key policy-  
12 relevant issues to be addressed in this review as a series of questions that frames our  
13 consideration of whether the current secondary (welfare-based) NAAQS for NO<sub>x</sub> and SO<sub>x</sub>  
14 should be retained or revised.

15 As part of this review, staff in the U.S. Environmental Protection Agency's (EPA) Office  
16 of Air Quality Planning and Standards (OAQPS) prepared this second draft Policy Assessment.<sup>1</sup>  
17 The objective of this assessment is to evaluate the policy implications of the key scientific  
18 information contained in the document *Integrated Science Assessment for Oxides of Nitrogen  
19 and Sulfur-Ecological Criteria* (USEPA, 2008; henceforth referred to as the ISA), prepared by  
20 EPA's National Center for Environmental Assessment (NCEA) and the results from the analyses  
21 contained in the *Risk and Exposure Assessment for Review of the Secondary National Ambient  
22 Air Quality Standards for Oxides of Nitrogen and Oxides of Sulfur* (U.S. EPA, 2009; henceforth  
23 referred to as the REA). This second draft also presents staff conclusions on a range of policy  
24 options that we believe are appropriate for the Administrator to consider concerning whether,  
25 and if so how, to revise the secondary (welfare-based) NO<sub>x</sub> and SO<sub>x</sub> NAAQS.

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<sup>1</sup> Preparation of a PA by OAQPS staff reflects Administrator Jackson's decision to modify the NAAQS review process that was presented in the IRP. See <http://www.epa.gov/ttn/naaqs/review.html> for more information on the current NAAQS review process.

1 This policy assessment is intended to help “bridge the gap” between the scientific  
2 assessment contained in the ISA and the judgments required of the EPA Administrator in  
3 determining whether it is appropriate to retain or revise the secondary NAAQS for NO<sub>x</sub> and  
4 SO<sub>x</sub>. This policy assessment considers the available scientific evidence and quantitative risk-  
5 based analyses, together with related limitations and uncertainties, and focuses on the basic  
6 components of air quality standards: indicators<sup>2</sup>, averaging times, forms<sup>3</sup>, and levels. These  
7 components, which serve to define each standard, must be considered collectively in evaluating  
8 the welfare protection afforded by the secondary NO<sub>x</sub> and SO<sub>x</sub> NAAQS. Our development of  
9 this policy assessment is based on the assessment and integrative synthesis of information  
10 presented in the ISA and on staff analyses and evaluations presented in this document, and is  
11 further informed by comments and advice received from an independent scientific review  
12 committee, the Clean Air Scientific Advisory Committee (CASAC), in their review of the  
13 previous integrated science assessment, risk and exposure assessment, and first draft policy  
14 assesment. To view related documents developed as part of the planning, science, and risk  
15 assessment phases of this review see

16 <http://www.epa.gov/ttn/naaqs/standards/no2so2sec/index.html>.

17 This document is organized around a conceptual framework for a combined NO<sub>x</sub> and SO<sub>x</sub>  
18 secondary NAAQS and is focused on answering key policy questions related to the  
19 implementation of that conceptual framework. Chapter 2 provides a summary of ecological  
20 effects from the deposition of ambient NO<sub>x</sub> and SO<sub>x</sub> to sensitive ecosystems, drawing from the  
21 ISA and REA. Chapter 3 places those ecological effects within the context of “public welfare”  
22 by linking effects to ecosystem services or other benchmarks of public welfare. Chapter 4  
23 addresses the adequacy of the current NO<sub>x</sub> and SO<sub>x</sub> secondary NAAQS in addressing the  
24 impacts on public welfare from ecological effects. Chapter 5 develops the conceptual design for  
25 ecologically relevant multi-pollutant standards and presents options for developing critical  
26 components of a secondary NAAQS necessary to implement the conceptual design. Chapter 6  
27 describes how secondary NAAQS designed to protect a specific ecological endpoint may also  
28 provide protection for other ecological endpoints. Chapter 7 provides an assessment of critical

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<sup>2</sup> The “indicator” of a standard defines the chemical species or mixture that is to be measured in determining whether an area attains the standard.

<sup>3</sup> The “form” of a standard defines the air quality statistic that is to be compared to the level of the standard in determining whether an area attains the standard.

1 uncertainties and sensitivities considered in developing the options for the components of the  
2 standard. Chapter 8 discusses issues related to monitoring of NO<sub>x</sub> and SO<sub>x</sub>. Chapter 9 provides  
3 initial staff conclusions regarding options for pollutant indicators, averaging times, forms, and  
4 ranges of levels for the secondary NO<sub>x</sub> and SO<sub>x</sub> NAAQS.

5 In this document we consider how the available scientific evidence and quantitative risk-  
6 based analyses, together with related limitations and uncertainties, inform the review of each  
7 element of the NAAQS: indicator, averaging times, forms, and levels. These components must be  
8 considered collectively in evaluating the welfare protection afforded by the secondary NAAQS  
9 standards. This draft document does not contain final staff conclusions as to all the necessary  
10 components of an alternative secondary standard for NO<sub>x</sub> and/or SO<sub>x</sub> but rather describes the  
11 current state of thinking with regard to potential policy options and provides an appropriate  
12 context of information for the Administrator to consider in making decisions regarding the  
13 standards.

14 While this policy assessment should be of use to all parties interested in the secondary  
15 NO<sub>x</sub> and SO<sub>x</sub> NAAQS review, it is written with an expectation that the reader has some  
16 familiarity with the technical discussions contained in the ISA and REA.

17 EPA's final Policy Assessment will address additional CASAC comments on this second  
18 draft, and will include sufficient information to inform the Administrator on critical components  
19 of the standards, and staff conclusions regarding alternative levels of the standards.

## 21 **1.1 DEFINITIONS, PARAMETERS, UNITS, AND CONVENTIONS USED FOR THIS** 22 **ASSESSMENT**

23 Throughout this document numerous terms are used that address a variety of atmospheric  
24 and ecosystem processes and variables. Some of the more common terms used in the technical  
25 community are not always synonymous with definitions imbedded in the CAA. Because of this  
26 diversity of terms spanning atmospheric and ecosystem processes along with adherence to  
27 scientific and legal conventions, this section provides the terminology as a reference source for  
28 the entire report.

29 As discussed in detail in the REA (**REA 1.3.1**), in the atmospheric science community  
30 NO<sub>x</sub> is typically referred to as the sum of nitrogen dioxide (NO<sub>2</sub>), and nitric oxide (NO). As  
31 defined by the Clean Air Act, the family of NO<sub>x</sub> includes any gaseous combination of nitrogen

1 and oxygen (e.g., NO<sub>2</sub>, NO, nitrous oxide [N<sub>2</sub>O], dinitrogen trioxide [N<sub>2</sub>O<sub>3</sub>], dinitrogen tetroxide  
2 [N<sub>2</sub>O<sub>4</sub>], and dinitrogen pentoxide [N<sub>2</sub>O<sub>5</sub>]). The term used by the scientific community to  
3 represent the complete set of reactive oxidized nitrogen compounds, including those listed in  
4 CAA Section 108(c) with the exception of N<sub>2</sub>O, is total oxidized nitrogen (NO<sub>y</sub>), commonly  
5 defined as NO, NO<sub>2</sub> and the all of the oxidation products of NO and NO<sub>2</sub>. Reactive oxidized  
6 nitrogen is defined as NO<sub>y</sub> = NO<sub>2</sub> + NO + HNO<sub>3</sub> + PAN + 2N<sub>2</sub>O<sub>5</sub> + HONO + NO<sub>3</sub> + organic  
7 nitrates + particulate NO<sub>3</sub> (Finlayson-Pitts and Pitts, 2000). In this document, unless otherwise  
8 indicated, we use the term NO<sub>y</sub> as the atmospheric indicators associated with the NO<sub>x</sub>  
9 component of the proposed NO<sub>x</sub>/SO<sub>x</sub> standard .

10 For this assessment, SO<sub>x</sub> is defined to include all oxides of sulfur, including multiple  
11 gaseous substances (e.g., SO<sub>2</sub>, sulfur monoxide [SO], sulfur trioxide [SO<sub>3</sub>], thiosulfate [S<sub>2</sub>O<sub>3</sub>],  
12 and heptoxide [S<sub>2</sub>O<sub>7</sub>], as well as particulate species, such as ammonium sulfate [(NH<sub>4</sub>)<sub>2</sub>SO<sub>4</sub>].  
13 Throughout this text we refer to sulfate as SO<sub>4</sub> and nitrate as NO<sub>3</sub>, recognizing that they have  
14 charges of -2 for sulfate and -1 for nitrate. The sum of sulfur dioxide gas (SO<sub>2</sub>) and particulate  
15 sulfate (SO<sub>4</sub>), referred herein as (SO<sub>2</sub> + SO<sub>4</sub>) is used throughout this document as the  
16 atmospheric indicator for the SO<sub>x</sub> component of the proposed NO<sub>x</sub>/SO<sub>x</sub> standard. From a  
17 measurement and modeling perspective we only consider the sum of SO<sub>2</sub> and particulate SO<sub>4</sub> as  
18 the indicator for sulfur. The sum of SO<sub>2</sub> and SO<sub>4</sub> constitute virtually all of the ambient air sulfur  
19 budget and are measured routinely in monitoring networks.

20 Table 1-1 provides further explanation of these indicators, some of which is repeated in  
21 Chapters 4, 5 and 8. Table 1 also provides details on the units used throughout the equations  
22 and examples in the PAD. Again, because of difference in unit conventions between  
23 atmospheric and ecosystem sciences, there are detailed explanations of units as well as  
24 procedures for translating between different unit conventions. To facilitate the linkage between  
25 atmospheric and ecosystem processes, only the mass (or equivalent charge) associated with  
26 sulfur or nitrogen is considered in mass, mixing ratio, and deposition unit conventions.

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1 **Table 1 -1. Description of parameters, units and conventions.**

Parameter	Units	Conversions to other unit conventions used in figures and calculations (multiply value in Units column by:	Explanation
<b>Atmospheric species</b>			
CMAQ defined NO <sub>y</sub> species: NO (nitrogen oxide), NO <sub>2</sub> (nitrogen dioxide), HNO <sub>3</sub> (nitric acid), p-NO <sub>3</sub> (particulate bound nitrate), NO <sub>3</sub> (sum of HNO <sub>3</sub> and p-NO <sub>3</sub> ), PAN (peroxy acetyl nitrate), N <sub>2</sub> O <sub>5</sub> (dinitrogen pentoxide), PANX (higher order PANs), NTR (organic nitrates), PNA (HNO <sub>3</sub> ); sulfur dioxide (SO <sub>2</sub> ), particulate sulfate (SO <sub>4</sub> ); NH <sub>x</sub> species: NH <sub>3</sub> (ammonia), ammonium ion (NH <sub>4</sub> )			
<b>Lumped Atmospheric Species</b>			
NO <sub>y</sub>	The sum of all reactive oxidized nitrogen compounds derived through summing all nitrogen contributions (i.e., 1·HNO <sub>3</sub> + 2·N <sub>2</sub> O <sub>5</sub> + ...) from the modeled species (HNO <sub>3</sub> , p-NO <sub>3</sub> , NO <sub>2</sub> , NO, PAN, ...) or through direct measurement which reduces all oxidized nitrogen species to NO and reports as ppb NO. <i>All references to the quantity NO<sub>y</sub> refer to the mass, molar or equivalent charge contribution of nitrogen only. All mass contributions of oxygen, hydrogen and carbon are not included.</i>		
(SO <sub>2</sub> +SO <sub>4</sub> )	Oxidized forms of sulfur defined as sulfate (SO <sub>4</sub> + SO <sub>2</sub> ); mass units maintained for consistency with deposition calculations <i>Note that only mass as sulfur is counted in state variables; in practice, individual SO<sub>2</sub> and SO<sub>4</sub> are measured/modeled and converted to mass of sulfur atoms or equivalent charge units. Mass contribution of oxygen is not included.</i>		
NH <sub>x</sub>	Reduced nitrogen calculated as the sum of NH <sub>3</sub> and NH <sub>4</sub> . <i>All references to the quantity NH<sub>x</sub> used as state variables refer to the mass, molar or equivalent charge contribution of nitrogen only. Mass contribution of oxygen is not included.</i>		
<b>Atmospheric State Variables used in equations and derivations</b>			
NO <sub>y</sub> concentration SO <sub>x</sub> NH <sub>x</sub>  Used in various conventions of:  C <sub>i</sub>	μg/m <sup>3</sup> as N or S	$ppb = (MA/M_i) \cdot \rho_{air} \cdot \mu g/m^3$  where ρ <sub>air</sub> is the air density in units of (kg/m <sup>3</sup> ); $\rho_{air} = 28.97 (10)^{-3} \cdot P/(R \cdot T)$ $R = 8.206(10)^{-5} m^3 atm/(mol \cdot K)$ $P = atm$ $T = degrees K$ MA = molecular weight of air (28.97) M <sub>i</sub> = Atomic weight of nitrogen (14) or sulfur (32) $meq/m^3 = (1/M_i) \cdot \mu g/m^3$	

Parameter	Units	Conversions to other unit conventions used in figures and calculations (multiply value in Units column by:	Explanation
$NO_y$ deposition $(SO_2 + SO_4)$ $NH_x$  Used in various conventions of: $Dep_i$	$meq/m^2\text{-yr}$ as N or S	$kg/ha\text{-yr} = (M_i/q)(10)^{-2} \cdot meq/m^2\text{-yr}$ where q = charge (1 for N, 2 for S)	
$C_{(SO_4+SO_2)}^{max}$	$\mu g/m^3$		is the concentration of $(SO_2 + SO_4)$ in the atmosphere consistent with $DL_S^{max}$
$C_{NO_y}^{max}$	$\mu g/m^3$		is the concentration of $NO_y$ in the atmosphere consistent with $DL_N^{max}$
$C_{NO_y}^{min}$	$\mu g/m^3$		is the concentration of $NO_y$ in the atmosphere consistent with $DL_N^{min}$
$v_i^{Dry}$	$m/yr$		dry deposition velocities
$v_i^{Wet}$	$m/yr$		wet deposition velocities
$Dep_i^{Dry}$	$meq/m^2\text{-yr}$		dry deposition fluxes
$Dep_i^{Wet}$	$meq/m^2\text{-yr}$		wet deposition fluxes
$Dep_i^{Total}$	$meq/m^2\text{-yr}$		total (wet+dry) deposition
$T_{SO_x}$ $T_{NO_y}$	$m/yr$	Calculated by dividing total $(SO_2 + SO_4)$ or $NO_y$ deposition (wet and dry) by the annual average $(SO_2 + SO_4)$ or $NO_y$ concentration.	the transfer ratio, which can be considered an aggregated, "effective" deposition velocity that relates total deposition of $(SO_2 + SO_4)$ or $NO_y$ to the total ambient concentration, and represents an average of the chemical species specific $v_i^{Tot}$ ( $= v_i^{Dry} + v_i^{Wet}$ ) values

Parameter	Units	Conversions to other unit conventions used in figures and calculations (multiply value in Units column by:	Explanation
<b>Ecosystem variables</b>			
ANC*	$\mu\text{eq/L}$		calculated value of ANC
ANC <sub>limit</sub>	$\mu\text{eq/L}$		a “target” ANC level ( $\mu\text{eq/L}$ )
CL <sub>ANClim(i)</sub>	$\text{meq/m}^2\text{-yr}$		depositional load that does not cause the catchment to exceed a given ANC <sub>lim</sub> , where <i>i</i> indicates the pollutant of interest
Q	$\text{m/yr}$		Average surface water runoff for an acid sensitive area (this is typically equal to precipitation – evapotranspiration)
Q <sub>ws</sub>	$\text{m/yr}$		Catchment level surface water runoff (m/yr) (this is typically equal to precipitation – evapotranspiration)
N <sub>ECO</sub>	$\text{meq/m}^2\text{-yr}$		nitrogen retention and denitrification by terrestrial catchment
N <sub>leach</sub>	$\text{meq/m}^2\text{-yr}$		N leaching
DL <sub>%eco(i)</sub>	$\text{meq/m}^2\text{-yr}$		The deposition metric, defined as the amount of deposition that protects a selected percentage of individual catchments for a population of water bodies from exceeding their DL <sub>ANClim(i)</sub> , where <i>i</i> indicates the pollutant of interest
DL <sub>N</sub> <sup>max</sup>	$\text{meq/m}^2\text{-yr}$		In the tradeoff curve for DL <sub>%eco(i)</sub> , the maximum of amount of N deposition when S deposition equals

Parameter	Units	Conversions to other unit conventions used in figures and calculations (multiply value in Units column by:	Explanation
			zero
$DL_N^{\min}$	$meq/m^2\text{-yr}$		In the tradeoff curve for $DL_{\%eco}(i)$ , the amount of N deposition that will be captured by the ecosystem before it leaches
$DL_S^{\max}$	$meq/m^2\text{-yr}$		In the tradeoff curve for $DL_{\%eco}(i)$ , the maximum amount of S sulfur deposition considering $N_{ECO}$

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## 2 1.2 POLICY OBJECTIVES

3 In conducting this periodic review of the NO<sub>x</sub> and SO<sub>x</sub> secondary NAAQS, EPA has  
4 decided to jointly assess the scientific information, associated risks, and standards relevant to  
5 protecting the public welfare from adverse effects associated with oxides of nitrogen and sulfur.  
6 Although EPA has historically adopted separate secondary standards for oxides of nitrogen  
7 (NO<sub>x</sub>) and oxides of sulfur (SO<sub>x</sub>), EPA is conducting a joint secondary review of these standards  
8 because NO<sub>x</sub>, SO<sub>x</sub>, and their associated transformation products are linked from an atmospheric  
9 chemistry perspective, as well as from an environmental effects perspective. The National  
10 Research Council (NRC) has recommended that EPA consider multiple pollutants, as  
11 appropriate, in forming the scientific basis for the NAAQS (NRC, 2004). There is a strong basis  
12 for considering these pollutants together, building upon EPA's and CASAC's past recognition of  
13 the interactions of these pollutants and on the growing body of scientific information that is now  
14 available related to these interactions and associated ecological effects.

15 EPA sets secondary standards for two criteria pollutants related to NO<sub>x</sub> and SO<sub>x</sub>: ozone  
16 and particulate matter (PM). NO<sub>x</sub> is a precursor to the formation of ozone in the atmosphere,  
17 and under certain conditions, can combine with atmospheric ammonia to form ammonium  
18 nitrate, a component of fine PM. SO<sub>x</sub> is a precursor to the formation of particulate sulfate,  
19 which is a significant component of fine PM in many parts of the U.S. While there are a number

1 of welfare effects associated with ozone and fine PM, including ozone damage to vegetation, and  
2 visibility degradation related to PM, protection against those effects is provided by the ozone and  
3 fine PM standards. This review focuses on evaluation of the protection provided by NO<sub>x</sub> and  
4 SO<sub>x</sub> secondary standards for effects associated with direct atmospheric concentrations of NO<sub>x</sub>  
5 and SO<sub>x</sub>, and effects associated with deposition of NO<sub>x</sub> and SO<sub>x</sub> to sensitive ecosystems,  
6 including deposition in the form of particulate nitrate and sulfate in their component forms.

7 The ISA highlights the ecological effects associated with deposition of ambient NO<sub>x</sub> and  
8 SO<sub>x</sub> to sensitive ecosystems other than commercially managed forests and agricultural lands.  
9 This assessment primarily focuses on the effects of ambient NO<sub>x</sub> and SO<sub>x</sub> via deposition on  
10 multiple ecological receptors, but also evaluates information on gas-phase effects of NO<sub>x</sub> and  
11 SO<sub>x</sub> via stomatal exposure on vegetation, which are the effects that the current secondary  
12 standards protect against. The ISA highlighted effects including those associated with  
13 acidification and nitrogen nutrient enrichment. Based on these highlighted effects, EPA's policy  
14 objective is to develop a framework for NO<sub>x</sub> and SO<sub>x</sub> standards that incorporate factors that will  
15 lead to standards that are ecologically relevant, and that recognizes the interactions between the  
16 two pollutants as they deposit to sensitive ecosystems, with an ultimate goal of setting standards  
17 that, based on the ecological criteria described in the ISA, and consistent with the requirements  
18 of the Clean Air Act, "are requisite to protect the public welfare from any known or anticipated  
19 adverse effects associated with the presence of such air pollutant in the ambient air."

20 In presenting policy options for the Administrator's consideration, we note that the final  
21 decision on retaining or revising the current secondary standards for NO<sub>x</sub> and SO<sub>x</sub> is largely a  
22 public welfare policy judgment based on the Administrator's informed assessment of what  
23 constitutes requisite protection against adverse effects to public welfare. A final decision should  
24 draw upon scientific information and analyses about welfare effects, exposure and risks, as well  
25 as judgments about the appropriate response to the range of uncertainties that are inherent in the  
26 scientific evidence and analyses. The ultimate determination as to what level of damage to  
27 ecosystems and the services provided by those ecosystems is adverse to public welfare is not  
28 wholly a scientific question, although it is informed by scientific studies linking ecosystem  
29 damage to losses in ecosystem services, and information on the value of those losses in  
30 ecosystem services. Our approach to informing these judgments, as discussed below, is  
31 consistent with the requirements of the NAAQS provisions of the Clean Air Act and with how

1 EPA and the courts have historically interpreted the Act. These provisions require the  
2 Administrator to establish secondary NAAQS that, in the Administrator’s judgment, are  
3 requisite to protect public welfare from any known or anticipated adverse effects associated with  
4 the presence of NOx and SOx in the ambient air. In so doing, the Administrator seeks to  
5 establish standards that are neither more nor less stringent than necessary for this purpose.

6 For this second draft policy assessment, we have chosen to focus much of our discussion  
7 on the effects of ambient NOx and SOx on ecological impacts associated with acidifying  
8 deposition of nitrogen and sulfur, which is a transformation product of ambient NOx and SOx.  
9 We have the greatest confidence in the causal linkages between NOx and SOx and aquatic  
10 acidification effects, and we have the most complete information available with which to develop  
11 an ecologically meaningful structure for the standards.

### 13 **1.3 CRITICAL POLICY ELEMENTS**

14 Our policy objective is guided by the information in the ISA and REA, framed within the  
15 legislative requirements of the CAA. This framing leads us to focus on critical policy elements  
16 (CPE) consistent with elements of Clean Air Act language.

17 Sections 108 and 109 of the CAA govern the establishment and periodic review of the  
18 NAAQS and of the air quality criteria upon which the standards are based. The NAAQS are  
19 established for pollutants that are listed under section 108, based on three criteria, including  
20 whether emissions of the air pollutant cause or contribute to air pollution which may reasonably  
21 be anticipated to endanger public health or welfare and whose presence in the ambient air results  
22 from numerous or diverse mobile or stationary sources. The NAAQS are based on air quality  
23 criteria that reflect the latest scientific knowledge, useful in indicating the types and extent of  
24 identifiable effects on public health or welfare that may be expected from the presence of the  
25 pollutant in ambient air. The criteria refer to criteria issued pursuant to §108 of the Clean Air  
26 Act, which include “(A) those variable factors (including atmospheric conditions) which of  
27 themselves or in combination with other factors may alter the effects on public health or welfare  
28 of such air pollutant; (B) the types of air pollutants which, when present in the atmosphere, may  
29 interact with such pollutant to produce an adverse effect on public health of welfare; and (C) any  
30 known or anticipated adverse effects on welfare.”

1 The following critical policy elements for the design of ecologically relevant secondary  
2 standards for NO<sub>x</sub> and SO<sub>x</sub> are identified:

3  
4 (CPE 1) An evaluation of the effects of ambient NO<sub>x</sub> and SO<sub>x</sub> on ecosystems, and the  
5 relationship between those effects and the measure of dose in the ecosystem,  
6 indicated by the depositional loadings of N and S.

7 (CPE 1.1) Evaluation of the relationship between response of ecological  
8 receptors, e.g. changes in diversity of fish species, and the response related to  
9 public welfare, e.g. loss in recreational fishing services.

10 (CPE 1.2) Evaluation of the extent to which identified effects are occurring  
11 under recent conditions, and the extent to which meeting the current standards  
12 would provide protection against these effects.

13  
14 (CPE 2) An assessment of how best to characterize, in defining the standards, the  
15 variable ecosystem factors that affect the relationship between ecological  
16 effects and depositional loadings of N and S.

17 (CPE 2.1) Specification of potential indicators of ecological effects, e.g. acid  
18 neutralizing capacity (ANC) that incorporates variability in ecosystem factors.

19  
20 (CPE 3) Characterization of the complex relationships between ambient concentrations  
21 of NO<sub>x</sub> and SO<sub>x</sub> and deposition of N and S in the specification of a standard.

22  
23 (CPE 4) Specification of the form for the standard(s), including ambient atmospheric  
24 indicators for NO<sub>x</sub> and SO<sub>x</sub>, with consideration of averaging times, and  
25 options for levels of the standard(s).

26  
27 The development of the conceptual framework for the NO<sub>x</sub> and SO<sub>x</sub> standards described  
28 in Section 1.4 will be motivated by these critical policy elements. However, in order to provide a  
29 historical context for this new framework, the next section provides a brief history of previous  
30 reviews of the NO<sub>x</sub> and SO<sub>x</sub> secondary NAAQS, as well as other relevant historical reviews of  
31 welfare effects associated with these pollutants.

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## **1.4 HISTORICAL CONTEXT**

### **1.4.1 History of NO<sub>x</sub> and SO<sub>x</sub> NAAQS Review**

#### **1.4.1.1 NO<sub>x</sub> NAAQS**

EPA began the most recent previous review of the NO<sub>x</sub> secondary standards in 1987 and in November 1991, EPA released an updated draft AQCD for CASAC and public review and comment (56 FR 59285). This draft document provided a comprehensive assessment of the available scientific and technical information on health and welfare effects associated with NO<sub>2</sub> and other NO<sub>x</sub>. CASAC reviewed the draft document at a meeting held on July 1, 1993, and concluded in a closure letter to the Administrator that the document “provides a scientifically balanced and defensible summary of current knowledge of the effects of this pollutant and provides an adequate basis for EPA to make a decision as to the appropriate NAAQS for NO<sub>2</sub>” (Wolff, 1993). The AQCD *Air Quality Criteria for Oxides of Nitrogen* was then finalized (U.S. EPA, 1993). EPA also prepared a Staff Paper that summarized and integrated the key studies and scientific evidence contained in the revised NO<sub>x</sub> AQCD and identified the critical elements to be considered in the review of the NO<sub>2</sub> NAAQS. CASAC reviewed two drafts of the Staff Paper and concluded in a closure letter to the Administrator that the document provided a “scientifically adequate basis for regulatory decisions on nitrogen dioxide” (Wolff, 1995). In October 1995, the Administrator announced her proposed decision not to revise either the primary or secondary NAAQS for NO<sub>2</sub> (60 FR 52874; October 11, 1995). A year later, the Administrator made a final determination not to revise the NAAQS for NO<sub>2</sub> after careful evaluation of the comments received on the proposal (61 FR 52852; October 8, 1996). The level for both the existing primary and secondary NAAQS for NO<sub>2</sub> is 0.053 ppm (100 micrograms per cubic meter [ $\mu\text{g}/\text{m}^3$ ] of air), annual arithmetic average, calculated as the arithmetic mean of the 1-hour NO<sub>2</sub> concentrations.

#### **1.4.1.2 SO<sub>x</sub> NAAQS**

Based on the 1970 SO<sub>x</sub> criteria document (DHEW, 1970), EPA promulgated primary and secondary NAAQS for SO<sub>2</sub> on April 30, 1971 (36 FR 8186). The secondary standards included a standard at 0.02 ppm in an annual arithmetic mean and a 3-hour average of 0.5 ppm, not to be exceeded more than once per year. These secondary standards were established solely on the basis of evidence of adverse effects on vegetation. In 1973, revisions made to Chapter 5

1 (“Effects of Sulfur Oxide in the Atmosphere on Vegetation”) of *Air Quality Criteria for Sulfur*  
2 *Oxides* (U.S. EPA, 1973) indicated that it could not properly be concluded that the vegetation  
3 injury reported resulted from the average SO<sub>2</sub> exposure over the growing season, rather than  
4 from short-term peak concentrations. Therefore, EPA proposed (38 FR 11355) and then finalized  
5 (38 FR 25678) a revocation of the annual mean secondary standard. At that time, EPA was aware  
6 that SO<sub>x</sub> air concentrations have other public welfare effects, including effects on materials,  
7 visibility, soils, and water. However, the available data were considered insufficient to establish  
8 a quantitative relationship between specific ambient SO<sub>x</sub> concentrations and effects (38 FR  
9 25679).

10 In 1979, EPA announced that it was revising the Air Quality Criteria Document (AQCD)  
11 for sulfur oxides concurrently with that for particulate matter and would produce a combined  
12 particulate matter and sulfur oxides criteria document. Following its review of a draft revised  
13 criteria document in August 1980, CASAC concluded that acid deposition was a topic of  
14 extreme scientific complexity because of the difficulty in establishing firm quantitative  
15 relationships among (1) emissions of relevant pollutants (e.g., SO<sub>2</sub> and oxides of nitrogen), (2)  
16 formation of acidic wet and dry deposition products, and (3) effects on terrestrial and aquatic  
17 ecosystems. CASAC also noted that acid deposition involves, at a minimum, several different  
18 criteria pollutants: oxides of sulfur, oxides of nitrogen, and the fine particulate fraction of  
19 suspended particles. CASAC felt that any document on this subject should address both wet and  
20 dry deposition, since dry deposition was believed to account for a substantial portion of the total  
21 acid deposition problem.

22 For these reasons, CASAC recommended that a separate, comprehensive document on  
23 acid deposition be prepared prior to any consideration of using the NAAQS as a regulatory  
24 mechanism for the control of acid deposition. CASAC also suggested that a discussion of acid  
25 deposition be included in the AQCDs for nitrogen oxides and PM and SO<sub>x</sub>. Following CASAC  
26 closure on the AQCD for SO<sub>2</sub> in December 1981, EPA’s Office of Air Quality Planning and  
27 Standards published a Staff Paper in November 1982, but the paper did not directly assess the  
28 issue of acid deposition. Instead, EPA subsequently prepared the following documents: *The*  
29 *Acidic Deposition Phenomenon and Its Effects: Critical Assessment Review Papers, Volumes I*  
30 *and II* (U.S. EPA, 1984a, b), and *The Acidic Deposition Phenomenon and Its Effects: Critical*  
31 *Assessment Document* (U.S. EPA, 1985) (53 FR 14935 -14936). These documents, though they

1 were not considered criteria documents and did not undergo CASAC review, represented the  
2 most comprehensive summary of relevant scientific information completed by EPA at that point.

3 On April 26, 1988 (53 FR 14926), EPA proposed not to revise the existing primary and  
4 secondary standards for SO<sub>2</sub>. This proposal regarding the secondary SO<sub>2</sub> NAAQS was due to the  
5 Administrator's conclusions that (1) based upon the then-current scientific understanding of the  
6 acid deposition problem, it would be premature and unwise to prescribe any regulatory control  
7 program at that time, and (2) when the fundamental scientific uncertainties had been decreased  
8 through ongoing research efforts, EPA would draft and support an appropriate set of control  
9 measures.

#### 11 **1.4.2 History of Related Assessments and Agency Actions**

12 In 1980, the Congress created the National Acid Precipitation Assessment Program  
13 (NAPAP) in response to growing concern about acidic deposition. The NAPAP was given a  
14 broad 10-year mandate to examine the causes and effects of acidic deposition and to explore  
15 alternative control options to alleviate acidic deposition and its effects. During the course of the  
16 program, the NAPAP issued a series of publicly available interim reports prior to the completion  
17 of a final report in 1990 (NAPAP, 1990).

18 In spite of the complexities and significant remaining uncertainties associated with the  
19 acid deposition problem, it soon became clear that a program to address acid deposition was  
20 needed. The Clean Air Act Amendments of 1990 included numerous separate provisions related  
21 to the acid deposition problem. The primary and most important of the provisions, the  
22 amendments to Title IV of the Act, established the Acid Rain Program to reduce emissions of  
23 SO<sub>2</sub> by 10 million tons and NO<sub>x</sub> emissions by 2 million tons from 1980 emission levels in order  
24 to achieve reductions over broad geographic regions. In this provision, Congress included a  
25 statement of findings that led them to take action, concluding that (1) the presence of acid  
26 compounds and their precursors in the atmosphere and in deposition from the atmosphere  
27 represents a threat to natural resources, ecosystems, materials, visibility, and public health; (2)  
28 the problem of acid deposition is of national and international significance; and (3) current and  
29 future generations of Americans will be adversely affected by delaying measures to remedy the  
30 problem.

1           Second, Congress authorized the continuation of the NAPAP in order to assure that the  
2 research and monitoring efforts already undertaken would continue to be coordinated and would  
3 provide the basis for an impartial assessment of the effectiveness of the Title IV program.

4           Third, Congress considered that further action might be necessary in the long term to  
5 address any problems remaining after implementation of the Title IV program and, reserving  
6 judgment on the form that action could take, included Section 404 of the 1990 Amendments  
7 (Clean Air Act Amendments of 1990, Pub. L. 101-549, § 404) requiring EPA to conduct a study  
8 on the feasibility and effectiveness of an acid deposition standard or standards to protect  
9 “sensitive and critically sensitive aquatic and terrestrial resources.” At the conclusion of the  
10 study, EPA was to submit a report to Congress. Five years later, EPA submitted its report,  
11 entitled *Acid Deposition Standard Feasibility Study: Report to Congress* (U.S. EPA, 1995) in  
12 fulfillment of this requirement. The Report concluded that establishing acid deposition standards  
13 for sulfur and nitrogen deposition may at some point in the future be technically feasible,  
14 although appropriate deposition loads for these acidifying chemicals could not be defined with  
15 reasonable certainty at that time.

16           Fourth, the 1990 Amendments also added new language to sections of the CAA  
17 pertaining to the scope and application of the secondary NAAQS designed to protect the public  
18 welfare. Specifically, the definition of “effects on welfare” in Section 302(h) was expanded to  
19 state that the welfare effects include effects “...whether caused by transformation, conversion, or  
20 combination with other air pollutants.”

21           In 1999, seven Northeastern states cited this amended language in Section 302(h) in a  
22 petition asking EPA to use its authority under the NAAQS program to promulgate secondary  
23 NAAQS for the criteria pollutants associated with the formation of acid rain. The petition stated  
24 that this language “clearly references the transformation of pollutants resulting in the inevitable  
25 formation of sulfate and nitrate aerosols and/or their ultimate environmental impacts as wet and  
26 dry deposition, clearly signaling Congressional intent that the welfare damage occasioned by  
27 sulfur and nitrogen oxides be addressed through the secondary standard provisions of Section  
28 109 of the Act.” The petition further stated that “recent federal studies, including the NAPAP  
29 Biennial Report to Congress: An Integrated Assessment, document the continued-and increasing-  
30 damage being inflicted by acid deposition to the lakes and forests of New York, New England  
31 and other parts of our nation, demonstrating that the Title IV program had proven insufficient.”

1 The petition also listed other adverse welfare effects associated with the transformation of these  
2 criteria pollutants, including impaired visibility, eutrophication of coastal estuaries, global  
3 warming, and tropospheric ozone and stratospheric ozone depletion.

4 In a related matter, the Office of the Secretary of the U.S. Department of Interior  
5 requested in 2000 that EPA initiate a rulemaking proceeding to enhance the air quality in  
6 national parks and wilderness areas in order to protect resources and values that are being  
7 adversely affected by air pollution. Included among the effects of concern identified in the  
8 request were the acidification of streams, surface waters, and/or soils; eutrophication of coastal  
9 waters; visibility impairment; and foliar injury from ozone.

10 In a Federal Register notice in 2001, EPA announced receipt of these requests and asked  
11 for comment on the issues raised in them. EPA stated that it would consider any relevant  
12 comments and information submitted, along with the information provided by the petitioners and  
13 DOI, before making any decision concerning a response to these requests for rulemaking (65 FR  
14 48699).

15 The most recent 2005 NAPAP report states that "... scientific studies indicate that the  
16 emission reductions achieved by Title IV are not sufficient to allow recovery of acid-sensitive  
17 ecosystems. Estimates from the literature of the scope of additional emission reductions that are  
18 necessary in order to protect acid-sensitive ecosystems range from approximately 40-80%  
19 beyond full implementation of Title IV.... The results of the modeling presented in this Report to  
20 Congress indicate that broader recovery is not predicted without additional emission reductions"  
21 (NAPAP, 2005).<sup>4</sup>

22 Given the state of the science as described in the ISA and in other recent reports, such as  
23 the NAPAP's above, EPA believes it is appropriate, in the context of evaluating the adequacy of  
24 the current NO<sub>2</sub> and SO<sub>2</sub> secondary standards in this review, to revisit the question of the  
25 appropriateness and the feasibility of setting a secondary NAAQS to address remaining known  
26 or anticipated adverse public welfare effects resulting from the acidic and nutrient deposition of  
27 these criteria pollutants.

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<sup>4</sup> Note that a new NAPAP report is expected to be released later in 2010. The findings of that report will be considered in the final policy assessment.

## 1 **1.5 PROPOSED CONCEPTUAL FRAMEWORK FOR COMBINED NO<sub>x</sub> SO<sub>x</sub>** 2 **STANDARDS**

3 There is a strong basis for considering NO<sub>x</sub> and SO<sub>x</sub> together at this time, building upon  
4 EPA's and CASAC's recognition of the interactions of these pollutants and on the growing body  
5 of scientific information that is now available related to these interactions and associated  
6 ecological effects. The REA introduced a conceptual framework for ecologically meaningful  
7 secondary standards that recognized the complex processes by which ecosystems are exposed to  
8 ambient NO<sub>x</sub> and SO<sub>x</sub>. That framework provided a flow from ambient concentrations exposures  
9 via deposition to ecological indicators and effects (see Figure ES-2 in the REA Executive  
10 Summary). This sequence represents the process by which we can determine the risks associated  
11 with ambient concentrations of NO<sub>x</sub> and SO<sub>x</sub>. However, for the purposes of discussing a  
12 conceptual framework for design of standards to protect against those risks, a modified version  
13 of the risk framework is needed.

14 **Figure 1-1** depicts the framework by which we are considering the structure of an  
15 ecologically meaningful secondary standard. It is a conceptual diagram that illustrates how a  
16 level of protection related to an indicator of ecological effect(s) equates to atmospheric  
17 concentrations of NO<sub>x</sub> and SO<sub>x</sub> indicators. This conceptual diagram illustrates the linkages  
18 between ambient air concentrations and resulting deposition metrics, and between the deposition  
19 metric and the ecological indicator of concern. The Atmospheric Deposition Transformation  
20 Function translates ambient atmospheric concentrations of NO<sub>x</sub> and SO<sub>x</sub> to nitrogen and sulfur  
21 deposition metrics, while the Ecological Effect Function transforms the deposition metric into  
22 the ecological indicator.

23 Development of a form for the standard that reflects this structure is a critical step in the  
24 overall standard setting process. The atmospheric levels of NO<sub>x</sub> and SO<sub>x</sub> that satisfy a particular  
25 level of ecosystem protection are those levels that result in an amount of deposition that is less  
26 than the amount of deposition that a given ecosystem can accept without defined levels of  
27 degradation of the ecological indicator for a targeted effect.

28 The details of this conceptual framework are discussed in Chapter 5, including  
29 discussions of modifying factors that alter the relationship between ambient atmospheric  
30 concentrations of NO<sub>x</sub> and SO<sub>x</sub> and depositional loads of nitrogen and sulfur, and those that  
31 modify the relationship between deposition loads and the ecological indicator.

1 In setting NAAQS to protect public health and welfare, EPA has historically established  
2 standards which require the comparison of monitored concentrations of an air pollutant against a  
3 numerical metric of atmospheric concentration that does not vary geographically. This approach  
4 has appropriately protected public health as at-risk populations are widely distributed throughout  
5 the nation. As more is learned about the effects of pollutants such as NO<sub>x</sub> and SO<sub>x</sub> and the  
6 environment, however, such an approach may not be appropriate to provide the requisite level of  
7 protection to public welfare from effects on sensitive ecosystems. EPA is proposing in this  
8 review of the secondary standard for NO<sub>x</sub> and SO<sub>x</sub> a standard that takes into account variable  
9 factors, such as atmospheric variables and location-specific characteristics of ecosystems, as the  
10 appropriate approach to protect the public welfare from the effects associated with the presence  
11 of these pollutants in the ambient air.

12 EPA must undertake a thorough review of the air quality criteria for the pollutant at issue  
13 in reviewing a secondary NAAQS, and determine whether a current standard is requisite to  
14 protect the public welfare. Under section 108 of the CAA, air quality criteria are to “reflect the  
15 latest scientific knowledge useful in indicating the kind and extent of all identifiable effects”  
16 associated with the presence of the pollutant in the ambient air. It is clear from the language of  
17 the CAA that where the state of the science provides a basis for considering such effects, the  
18 review of the air quality criteria should encompass a broad analysis of “any” known or  
19 anticipated adverse effects, as well as the ways in which variable conditions such as atmospheric  
20 conditions may impact the effect of a pollutant and the ways in which other air pollutants may  
21 interact with the criteria pollutant to produce adverse effects. Specifically, section 108(a)(2) of  
22 the CAA provides that:

23 Air quality criteria for an air pollutant shall accurately reflect the latest scientific  
24 knowledge useful in indicating the kind and extent of all identifiable effects on public health or  
25 welfare which may be expected from the presence of such pollutant in the ambient air, in varying  
26 quantities. The criteria for an air pollutant to the extent practicable, shall include information on:

- 27 (A) Those variable factors (including atmospheric conditions) which of themselves or  
28 in combination with other factors may alter the effects on public health or welfare  
29 of such air pollutants;
- 30 (B) The types of air pollutants which, when present in the atmosphere, may interact  
31 with such pollutants to produce an adverse effect on public health or welfare; and

1 (C) Any known or anticipated adverse effects on welfare.

2  
3 Based on this extensive review of the air quality criteria for an air pollutant, the  
4 Administrator is required to review and to revise, as appropriate, the secondary standard to  
5 ensure that the standard “is requisite to protect public welfare from any known or anticipated  
6 adverse effects associated with the presence of such air pollutant in the ambient air.” CAA §  
7 109(b) & (d). “Effects on welfare,” in turn, is defined to include a broad array of effects,  
8 including effects on soil, water, crops, vegetation, and manmade materials, “whether caused by  
9 transformation, conversion, or combination with other air pollutants.” CAA § 302(h). Thus, as  
10 with the sections of the CAA describing the issuance of air quality criteria, the CAA uses  
11 expansive language in describing the scope of EPA’s responsibility and the range of effects that  
12 EPA should take into account in setting a standard that is requisite to protect public welfare. The  
13 term “requisite,” however, indicates that section 109 is not open-ended. In considering the  
14 meaning of the term “requisite” in the context of the primary standards, the Supreme Court has  
15 agreed with EPA that such a standard is one that is “sufficient, but not more than necessary” to  
16 protect public health. *Whitman v. American Trucking*, 531 U.S. 457, 473 (2001).

17 While EPA has most often considered the results of direct exposure to an air pollutant in  
18 the ambient air in assessing effects on public health and welfare, such as the health effects on  
19 humans when breathing in an air pollutant or the effects on vegetation through the uptake of air  
20 pollutants from the ambient air through leaves, EPA has also considered, where appropriate, the  
21 effects of exposure to air pollutants through more indirect mechanisms. For example, both in  
22 1978 and in 2008, EPA established a NAAQS for lead that addressed the health effects of  
23 ambient lead whether the lead particles were inhaled or were ingested after deposition on the  
24 ground or other surfaces. 73 FR 66964 (November 12, 2008), *Lead Industries v. EPA*, 647 F.2d  
25 1130 (DC Cir. 1980) (1978 NAAQS). The deposition of ambient NO<sub>x</sub> and SO<sub>x</sub> to terrestrial and  
26 aquatic environments can impact ecosystems through both direct and indirect mechanisms, as  
27 discussed in the REA and this document. Given Congress’ instruction to set a standard that “is  
28 requisite to protect the public welfare from “any known or anticipated adverse effects associated  
29 with the presence of such air pollutant in the ambient air,” 42 U.S.C. § 109 (b)(2), this review  
30 appropriately attempts to take into consideration widely acknowledged effects, such as

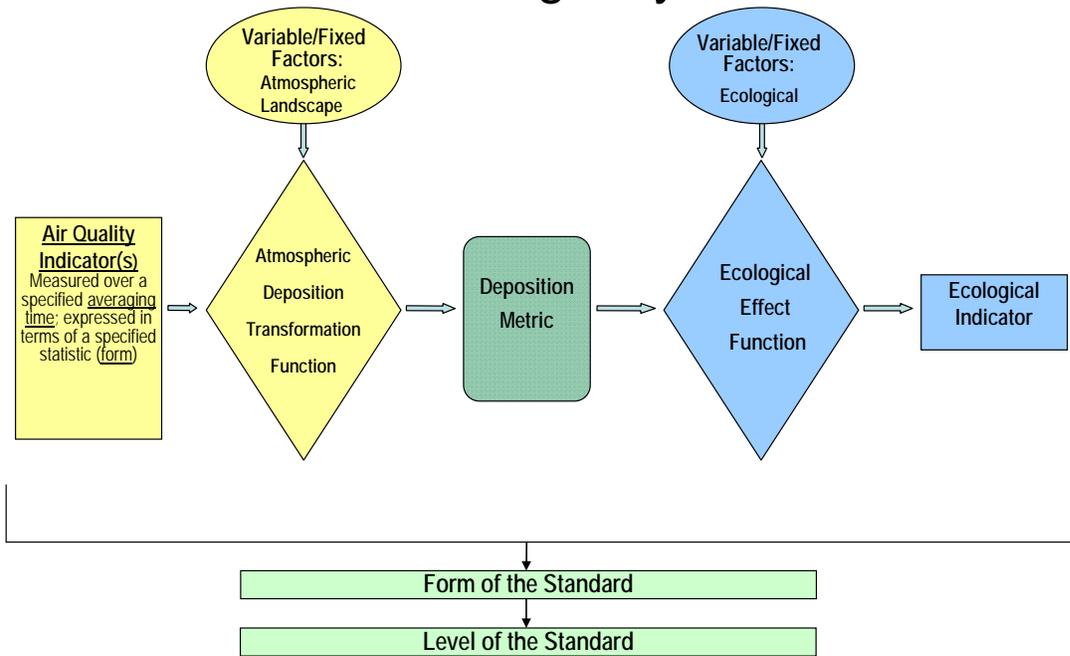
1 acidification and nutrient enrichment, which are associated with the presence of ambient SOx  
2 and NOx.

3 In this review, EPA is also attempting to develop a standard that takes into account the  
4 variability in effects from ambient levels of SOx and NOx. The CAA requires EPA to establish  
5 “national” standards, based on the air quality criteria that provide the requisite degree of  
6 protection, but does not clearly address how to do so under the circumstances present here. One  
7 approach is to develop a secondary standard such as the one discussed in this Policy Assessment  
8 Document. Such a standard is designed to provide a generally uniform degree of protection  
9 throughout the country by allowing for varying concentrations of allowable ambient NOx and  
10 SOx, depending on atmospheric conditions and other variabilities, to achieve that degree of  
11 protection<sup>5</sup>. Such a standard protects sensitive ecosystems wherever such ecosystems are found.  
12 This approach recognizes that setting a standard that is sufficient to protect the public welfare but  
13 not more than is necessary calls for consideration of a standard such as the one discussed in this  
14 document.

---

<sup>5</sup> In concept, this approach to setting a national standard using a consistent form that may result in differing levels of atmospheric concentrations of NOx and SOx is similar to the current proposal to use PM10 as the national indicator for protection against the health effects of PM10-2.5, as discussed in the 2010 2<sup>nd</sup> draft Policy Assessment for the PM NAAQS primary standards. In that case, EPA is proposing to recognize that the same size fraction of particles may have different toxicity depending on location, and as a result, it is more appropriate to use an indicator that reflects that varying toxicity, rather than setting one absolute level of PM10-2.5 which may not be equally protective in all locations. By proposing this form, EPA is recognizing that in attaining a PM10 standard, the resulting balance of PM10-2.5 and PM2.5 particles will differ across areas of the U.S. Likewise, setting a joint NOx/SOx standard that results in differing allowable concentrations of NOx and SOx across the U.S. based on the differing potential for NOx and SOx to result in ecological damages is appropriate in providing a requisite level of welfare protection.

# Structure of an Ecologically-based Standard



1  
2  
3 **Figure 1-1. Framework of an alternative secondary standard.**

4  
5 **1.6 POLICY RELEVANT QUESTIONS**

6 In this policy assessment, a series of general questions frames our approach to identifying  
7 a range of policy options for consideration by the Administrator regarding secondary NAAQS  
8 for NO<sub>x</sub> and SO<sub>x</sub>. These questions are drawn from our Integrated Review Plan with  
9 modifications based on further consideration by staff and comments from CASAC and the  
10 public. Our policy assessment begins by characterizing “known or anticipated adverse effects”  
11 on public welfare within our conceptual model (CPE 1). As noted earlier, this review is focusing  
12 on effects in sensitive unmanaged ecosystems (not commercial forests or agricultural lands<sup>6</sup>)  
13 resulting from ambient concentrations of NO<sub>x</sub> and SO<sub>x</sub> through deposition of N and S.

---

<sup>6</sup> The decision to focus on unmanaged ecosystems is based on the weight of evidence of effects in those ecosystems. The majority of the scientific evidence regarding acidification and nutrient enrichment is based on studies in unmanaged ecosystems. Non-managed terrestrial ecosystems tend to have a higher fraction of N deposition resulting from atmospheric N (ISA 3.3.2.5). In addition, the ISA notes that agricultural and commercial forest lands are routinely fertilized with amounts of N (100 to 300 kg N/ha) that exceed air pollutant inputs even in the most polluted areas (ISA 3.3.9). This review recognizes that effect of N deposition in managed areas may be viewed differently than effects of N deposition in unmanaged ecosystems, largely due to the more homogeneous, controlled nature of species composition and development in managed ecosystems and the potential for benefits of increased productivity in those ecosystems.

1 In Chapter 2, we draw from the information and conclusions presented in the ISA and  
2 REA to address the following questions:

- 3 1. What are the nature and magnitude of ecosystem responses to reactive nitrogen and  
4 sulfur deposition, acidification, nutrient depletion and the mobilization of toxic metals  
5 in sensitive aquatic and terrestrial ecosystems?
  - 6 a. How are these responses affected by landscape factors?
  - 7 b. What types of ecosystems are sensitive to such responses?
- 8 2. To what extent can ecosystem responses to nitrogen deposition be separated into  
9 responses related to oxidized and reduced forms of reactive nitrogen compounds?

10  
11 In Chapter 3, we address the following questions related to linking effects to measures of  
12 adversity (CPE 1.1):

- 13 1. How do we characterize adversity to public welfare? What are the sources of  
14 potentially relevant characterization for this policy assessment?
- 15 2. What is the evidence of effects on ecosystem services, and how can those ecosystem  
16 services be linked to ecological indicators?
- 17 3. To what extent are identified ecosystem effects important from a public welfare  
18 perspective, and what are the important uncertainties associated with estimating such  
19 effects?

20  
21 Once we have described ecological effects, we then provide an assessment of the  
22 adequacy of the existing NO<sub>x</sub> and SO<sub>x</sub> standards (CPE 1.2). We begin this assessment by  
23 drawing from the information and conclusions presented in the ISA and REA to address in  
24 Chapter 4 the following questions, which allow us to identify whether the structure of the current  
25 standards is appropriate relative to the key ecological effects assessed in the ISA and REA,  
26 including acidification and excess nutrient enrichment and whether there is adequate information  
27 and analyses available at this time to assess the extent to which potentially adverse effects on  
28 aquatic and terrestrial ecosystems can be associated with current levels of atmospheric reactive  
29 nitrogen, accounting for the contributions of oxidized and reduced forms, and SO<sub>x</sub> and with  
30 levels that are at or below the current secondary standards:

- 1 1. To what extent are effects that could reasonably be judged to be adverse to public  
2 welfare occurring under current conditions and would such effects occur if the nation  
3 met the current standards? To what extent do the current NO<sub>x</sub> and SO<sub>x</sub> secondary  
4 standards provide protection from effects associated with deposition of:
  - 5 a. Sulfur and oxidized nitrogen from atmospheric NO<sub>x</sub>, and SO<sub>x</sub> which results in  
6 acidification in sensitive aquatic and terrestrial ecosystems?
  - 7 b. Oxidized nitrogen from atmospheric NO<sub>x</sub>, which results in nutrient enrichment  
8 effects in sensitive aquatic and terrestrial ecosystems?
  - 9 c. Sulfur and oxidized nitrogen from atmospheric NO<sub>x</sub> and SO<sub>x</sub> which results in  
10 other ecological effects (e.g. mercury methylation)?
- 11 2. In what way are the structures of the current NO<sub>x</sub> and SO<sub>x</sub> secondary standards  
12 inadequate to protect against public welfare effects?  
13

14 In Chapter 5, we follow our adequacy assessment by developing in greater detail the  
15 conceptual framework for the design of ecologically relevant multi-pollutant standards  
16 introduced in Section 1.4 above. To the extent that the available information calls into question  
17 the adequacy of protection afforded by the current standards and/or the appropriateness of the  
18 structure of the standards, we explore the extent to which available information supports  
19 consideration of alternative standards, in terms of atmospheric and ecological indicators and  
20 related averaging times, forms, and levels. This conceptual framework is designed to focus on  
21 resolving the following questions:

- 22 1. (CPE 2.1) Does the available information provide support for the use of ecological  
23 indicators to characterize the responses of aquatic and terrestrial ecosystems to  
24 oxidized nitrogen and sulfur deposition?
- 25 2. (CPE 1) Does the available information provide support for the development of  
26 appropriate ecological response to deposition relationship(s) that meaningfully relates  
27 oxidized nitrogen and sulfur deposition to relevant ecological indicators? Does a  
28 quantified relationship exist between the level of a relevant ecological indicator and  
29 an amount of nitrogen and sulfur deposition?

- 1 3. (CPE 2) What are the important variables in the ecological response to deposition  
2 relationship(s)? Are these relationships applicable nationally? What are the  
3 appropriate temporal scales for these relationships?
  - 4 a. How does ecological response to deposition relationship(s) depend upon  
5 spatially heterogeneous geologic factors (e.g. bedrock type, weathering rates)  
6 that govern sensitivity?
  - 7 b. How do we consider areas with high natural background acidification or  
8 nutrient loadings?
- 9 4. (CPE 3) Does the available information provide support for the development of  
10 appropriate functions that characterize the relationships between atmospheric NO<sub>x</sub>  
11 and SO<sub>x</sub> and the wet and dry deposition of total reactive nitrogen and sulfur?
  - 12 a. What deposition function is appropriate to use for the purpose of relating an  
13 amount of nitrogen and/or sulfur deposition in sensitive ecosystems to  
14 ambient concentrations of atmospheric reactive nitrogen, including oxides and  
15 reduced forms, and/or sulfur? What are the important variables in such a  
16 function? What are appropriate spatial and temporal scales to use in  
17 specifying such variables?

18  
19 Based on the conceptual framework for the structure of the ecologically relevant multi-  
20 pollutant standards, we then address in Chapter 6 the components of the standard needed to  
21 develop options for consideration by the Administrator. Development of these options will focus  
22 on addressing the following questions:

- 23 1. (CPE 2.1) What ecological indicators are appropriate to use for the purpose of  
24 developing an alternative standard for the various ecological effects assessed in this  
25 review?
- 26 2. (CPE 5) What indicators of oxides of nitrogen and sulfur are appropriate to use for  
27 the purpose of determining whether the resultant deposition is within the target values  
28 needed to achieve the desired degree of protection? What averaging times and forms  
29 are appropriate to consider?

- 1 3. (CPE 2) What approaches are available to specify non-atmospheric components of  
2 the standard, e.g. weathering rates? Are there approaches that can simplify the  
3 structure of the standard?
- 4 4. What are the available approaches for accounting for reduced N in the structure of the  
5 standard?
- 6 5. What is the most appropriate form for the standards to reflect the relationships  
7 between ambient NO<sub>x</sub> and SO<sub>x</sub>, acidifying deposition, and the ecological indicator  
8 for acidification?

9 Several follow-up questions derive from our assessment of options for specifying the  
10 components of a multipollutant standard. In Chapter 6, we address the questions:

- 11 1. To what extent would a standard specifically defined to protect against one ecological  
12 effect (i.e., aquatic acidification) likely provide protection from other relevant  
13 ecological effects?
- 14 2. What are the available approaches for combining multiple indicators into a single  
15 standard, e.g. using nitrogen effects to bound the tradeoff curve for NO<sub>x</sub>/SO<sub>x</sub> for  
16 aquatic acidification effects
- 17 3. What are the available approaches to integrate potential standards for aquatic and  
18 terrestrial acidification and/or aquatic and terrestrial N enrichment?

19  
20 In Chapter 7, we provide a range of explorations of uncertainties in the evidence and  
21 models as they pertain to the selection of options for components of the standard. In addition,  
22 we provide results of sensitivity analyses for components of the proposed AAPI form, as well as  
23 characterizing information on variability in those components. The chapter focuses on the  
24 following questions:

- 25 1. What are critical uncertainties in the characterization of pre-industrial levels of  
26 ANC?
- 27 2. What uncertainties are introduced through the use of steady state critical load  
28 models relative to dynamic critical loads models?
- 29 3. What are the critical uncertainties in the modeled relationship between  
30 concentrations of NO<sub>x</sub> and SO<sub>x</sub> and deposition of N and S?
- 31 4. What are the critical uncertainties in the modeled values of NH<sub>x</sub> deposition?

- 1           5.     How sensitive is the form of the NO<sub>x</sub>/SO<sub>x</sub> standard to the primary components?
- 2           6.     How sensitive are the atmospheric transformation ratios calculated from CMAQ
- 3                 to the emissions scenarios, chemical mechanisms, and meteorology in the model?
- 4           7.     How well does CMAQ perform in simulating nitric oxide, nitrogen dioxide, sulfur
- 5                 dioxide, nitrate, ammonium and aerosol nitrate, ammonium, and sulfate relative to
- 6                 observations from different networks for which the data are routinely available?
- 7           8.     How well does CMAQ perform in simulating wet deposition of sulfate, nitrate,
- 8                 and ammonium relative to observations from the National Atmospheric
- 9                 Deposition Program (NADP) network?

10  
11           Chapter 8 provides a discussion of several important aspects of monitoring of NO<sub>x</sub> and

12 SO<sub>x</sub>, including methods, network design, and frequency.

13           We conclude in Chapter 9 with a discussion of options to consider in selecting pollutant

14 indicators, averaging times, forms, and ranges of levels for the secondary NO<sub>x</sub> and SO<sub>x</sub>

15 standards. This discussion is informed by a consideration of the role of ecosystem services in

16 helping to characterize what adversity to public welfare, focused on the following questions:

- 17           1. (CPE 5) What are the risks of ecosystem service impairment under alternative levels
- 18                 of potential standards for NO<sub>x</sub> and SO<sub>x</sub>?
- 19           2. (CPE 5) To what extent can information about ecosystem services be used to help
- 20                 characterize the extent to which differing levels of relevant ecological indicators
- 21                 reflect impacts that can reasonably be judged to be adverse from a public welfare
- 22                 perspective?
- 23           3. (CPE 5) Are there relevant benchmarks for adversity to public welfare that can be
- 24                 derived from other sources?
- 25           4. (CPE 5) Taking into consideration information about ecosystem services and other
- 26                 factors related to characterizing adversity to public welfare for the ecological effects
- 27                 being assessed in this review, what is an appropriate range of levels of protection to
- 28                 be achieved by alternative standards for the Agency to consider?

## 2 KNOWN OR ANTICIPATED ECOLOGICAL EFFECTS

In this chapter we address Critical Policy Element 1, evaluation of the effects of ambient NO<sub>x</sub> and SO<sub>x</sub> on ecosystems, and the relationship between those effects and the measure of dose in the ecosystem, indicated by the depositional loadings of N and S. In section 302(h) of the Clean Air Act, welfare effects addressed by a secondary NAAQS include, but are not limited to, “effects on soils, water, crops, vegetation, man-made materials, animals, wildlife, weather, visibility and climate, damage to and deterioration of property, and hazards to transportation, as well as effects on economic values and on personal comfort and well-being”. Of these welfare effects categories, the effects of NO<sub>x</sub> and SO<sub>x</sub> on aquatic and terrestrial ecosystems, which encompass soils, water, vegetation, wildlife, and contribute to economic value and well-being, are of most concern at concentrations typically occurring in the U.S. Direct effects of NO<sub>x</sub> and SO<sub>x</sub> on vegetation are also discussed in this chapter, and have been the focus of previous reviews. However, for this review, the focus of this chapter is on the known and anticipated effects to ecosystems caused by exposure to NO<sub>x</sub> and SO<sub>x</sub> through deposition.

The information presented here is a concise summary of conclusions from the ISA and the REA. This chapter focuses on effects on specific ecosystems with a brief discussion on critical uncertainties associated with acidification and nutrient enrichment. Those effects are then evaluated in Chapter 3 within the context of alternative definitions of, including assessments of potential impacts on ecosystem services. Effects are broadly categorized into acidification and nutrient-enrichment in the proceeding sections. This is background information intended to support new approaches for the design of ecologically relevant secondary NO<sub>x</sub> and SO<sub>x</sub> standards which are protective of U.S. ecosystems. More detailed information on the conceptual design and specific options for the proposed standards are presented in Chapters 5 and 9 of this policy assessment document. While we provide a summary of effects for all four of the primary effects categories, we reiterate that the focus of this second draft policy assessment is on effects related to aquatic acidification, without downplaying the potential importance of effects in other categories.

1 **2.1 ACIDIFICATION: EVIDENCE OF EFFECTS ON STRUCTURE AND**  
2 **FUNCTION OF TERRESTRIAL AND FRESHWATER ECOSYSTEMS**

3 Sulfur oxides (SO<sub>x</sub>) and nitrogen oxides (NO<sub>x</sub>) in the atmosphere undergo a complex mix of  
4 reactions in gaseous, liquid, and solid phases to form various acidic compounds. These acidic  
5 compounds are removed from the atmosphere through deposition: either wet (e.g., rain, snow),  
6 fog or cloud, or dry (e.g., gases, particles). Deposition of these acidic compounds to ecosystems  
7 can lead to effects on ecosystem structure and function. Following deposition, these compounds  
8 can, in some instances unless buffered by high base soils, leach out of the soils in the form of  
9 sulfate (SO<sub>4</sub><sup>2-</sup>) and nitrate (NO<sub>3</sub><sup>-</sup>), leading to the acidification of surface waters. The effects on  
10 ecosystems depend on the magnitude and rate of deposition, as well as a host of biogeochemical  
11 processes occurring in the soils and waterbodies (**REA 2.1**). The chemical forms of nitrogen that  
12 may contribute to acidifying deposition include both oxidized and reduced chemical species.

13 When sulfur or nitrogen (NO<sub>x</sub>, NH<sub>x</sub> and Nr) leaches from soils to surface waters in the  
14 form of SO<sub>4</sub><sup>2-</sup> or NO<sub>3</sub><sup>-</sup>, an equivalent amount of positive cations, or countercharge, is also  
15 transported. This maintains electroneutrality. If the countercharge is provided by base cations,  
16 such as calcium (Ca<sup>2+</sup>), magnesium (Mg<sup>2+</sup>), sodium (Na<sup>+</sup>), or potassium (K<sup>+</sup>), rather than hydrogen  
17 (H<sup>+</sup>) and dissolved inorganic aluminum, the acidity of the soil water is neutralized, but the base  
18 saturation of the soil decreases. Continued SO<sub>4</sub><sup>2-</sup> or NO<sub>3</sub><sup>-</sup> leaching can deplete the available base  
19 cation pool in soil. As the base cations are removed, continued deposition and leaching of SO<sub>4</sub><sup>2-</sup>  
20 and/or NO<sub>3</sub><sup>-</sup> (with H<sup>+</sup> and Al<sup>3+</sup>) leads to acidification of soil water, and by connection, surface  
21 water. The ability of a watershed to neutralize acidic deposition is determined by a variety of  
22 biogeophysical factors including weathering rates, bedrock composition, vegetation and  
23 microbial processes, physical and chemical characteristics of soils and hydrologic flowpaths.  
24 (**REA 2.1**) Some of these factors such as vegetation and soil depth are highly variable over  
25 small spatial scales such as meters, but can be aggregated to evaluate patterns over larger spatial  
26 scales. For the purpose of a national secondary standard, the most relevant characteristics are  
27 those that are less variable over small scales.

28 Acidifying deposition of NO<sub>x</sub> and SO<sub>x</sub> and the chemical and biological responses  
29 associated with these inputs vary temporally. Chronic or long-term deposition processes in the  
30 time scale of years to decades result in increases in inputs of N and S to ecosystems and the  
31 associated ecological effects. Episodic or short term (i.e., hours or days) deposition refers to

1 events in which the level of the acid neutralizing capacity (ANC) of a lake or stream is  
2 temporarily lowered. In aquatic ecosystems, short-term (i.e., hours or days) episodic changes in  
3 water chemistry can have significant biological effects. Episodic acidification refers to  
4 conditions during precipitation or snowmelt events when proportionately more drainage water is  
5 routed through upper soil horizons that tend to provide less acid neutralizing than was passing  
6 through deeper soil horizons (**REA 4.2**). Some streams and lakes may have chronic or base flow  
7 chemistry that is suitable for aquatic biota, but may be subject to occasional acidic episodes with  
8 deleterious consequences to sensitive biota.

9 The following summary is a concise overview of the known or anticipated effects caused  
10 by acidification to ecosystems within the United States. Acidification affects both terrestrial and  
11 freshwater aquatic ecosystems. Terrestrial and aquatic processes are often linked; therefore  
12 responses to the following questions address both types of ecosystems unless otherwise noted.

13

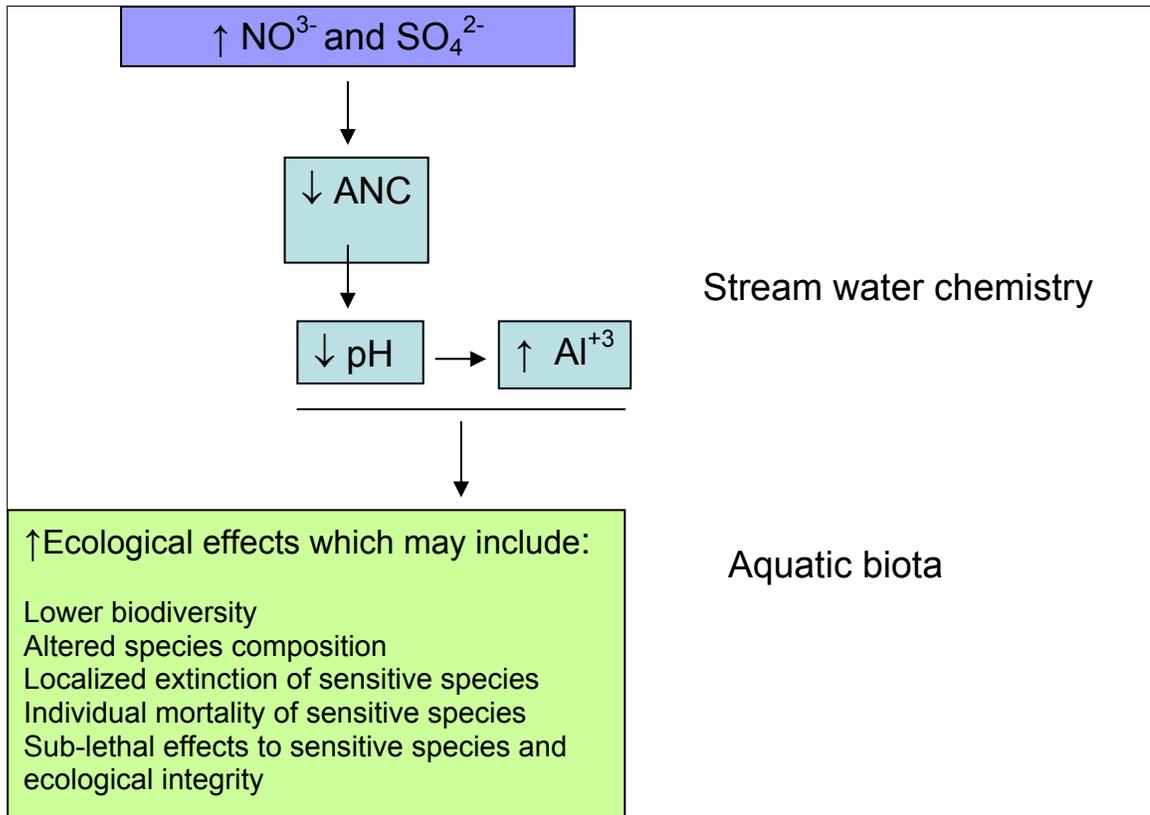
#### 14 **2.1.1 What is the nature of acidification related ecosystem responses to reactive** 15 **nitrogen and sulfur deposition?**

16 The ISA concluded that deposition of SO<sub>x</sub>, NO<sub>x</sub>, and NH<sub>x</sub> leads to the varying degrees of  
17 acidification of ecosystems (EPA 2008). In the process of acidification, biogeochemical  
18 components of terrestrial and freshwater aquatic ecosystems are altered in a way that leads to  
19 effects on biological organisms. Deposition to terrestrial ecosystems often moves through the  
20 soil and eventually leaches into adjacent water bodies.

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

1 biological effects are caused by lowered pH and increased inorganic Al concentrations. While  
 2 ANC level does not cause direct biological harm it is a good overall indicator of the risk of  
 3 acidification (Fig 2-1, See further discussion in Section 2.1.3).

4



5

6

7 **Figure 2-1. Conceptual model of direct and indirect acidification effects on aquatic biota.**  
 8 **Acidic pollutants (NO<sub>3</sub><sup>-</sup> and SO<sub>4</sub><sup>2-</sup>) lower ANC, resulting in lower pH with**  
 9 **direct toxic effects on fish. The lower pH mobilizes Al<sup>3+</sup> from soils often**  
 10 **resulting in higher concentration in stream water causing direct toxicity to**  
 11 **fish.**

12

13 These changes in stream water chemistry contribute to declines in taxonomic richness of  
 14 zooplankton, macroinvertebrates, and fish, which are often sources of food for birds and other  
 15 animal species in various ecosystems. These fish may also serve as a source of food and  
 16 recreation for humans (see Chapter 3). Acidification of ecosystems has been shown to disrupt  
 17 food web dynamics causing alteration to the diet, breeding distribution and reproduction of  
 18 certain species of birds (**ISA Section 4.2.2.2. and Table 3-9**). For example, breeding  
 19 distribution of the common goldeneye (*Bucephala clangula*) an insectivorous duck, may be

1 affected by changes in acidifying deposition (Longcore and Gill, 1993). Similarly, decreases in  
2 prey diversity and quantity have been observed to create feeding problems for nesting pairs of  
3 loons on low-pH lakes in the Adirondacks (Parker 1988).

4 *In terrestrial ecosystems, the evidence is sufficient to infer a **causal** relationship between*  
5 *acidifying deposition and changes in biogeochemistry (ISA 4.2.1.1).* The strongest evidence  
6 comes from studies of forested ecosystems, with supportive information on other plant taxa,  
7 including shrubs and lichens (ISA 3.2.2.1.). Three useful indicators of chemical changes and  
8 acidification effects on terrestrial ecosystems, showing consistency and coherence among  
9 multiple studies are: soil base saturation, Al concentrations in soil water and soil C:N ratio (ISA  
10 3.2.2.2).

11 In soils with base saturation less than about 15 to 20%, exchange chemistry is dominated  
12 by Al (Reuss, 1983). Under these conditions, responses to inputs of sulfuric acid and nitric acid  
13 largely involve the release and mobilization of dissolved inorganic Al. The effect can be  
14 neutralized by weathering from geologic parent material or base cation exchange. The Ca<sup>2+</sup> and  
15 Al concentrations in soil water are strongly influenced by soil acidification and both have been  
16 shown to have quantitative links to tree health, including Al interference with Ca<sup>2+</sup> uptake and Al  
17 toxicity to roots (Parker et al., 1989; U.S. EPA, 2009). Effects of nitrification and associated  
18 acidification and cation leaching have been consistently shown to occur only in soils with a C:N  
19 ratio below about 20 to 25 (Aber et al., 2003; Ross et al., 2004).

20 Soil acidification caused by acidic deposition has been shown to cause decreased growth  
21 and increased susceptibility to disease and injury in sensitive tree species. Red spruce (*Picea*  
22 *rubens*) dieback or decline has been observed across high elevation areas in the Adirondack,  
23 Green and White mountains (DeHayes et al., 1999). The frequency of freezing injury to red  
24 spruce needles has increased over the past 40 years, a period that coincided with increased  
25 emissions of S and N oxides and increased acidifying deposition (DeHayes et al., 1999).  
26 Acidifying deposition can contribute to dieback in sugar maple (*Acer saccharum*) through  
27 depletion of cations from soil with low levels of available Ca (Horsley et al., 2000; Bailey et al.,  
28 2004). Grasslands are likely less sensitive to acidification than forests due to grassland soils  
29 being generally rich in base cations (Fenn et al., 2003; Blake et al., 1999).

1 **2.1.2 What types of ecosystems are sensitive to such effects? In which ways are these**  
2 **responses affected by atmospheric, ecological, and landscape factors?**

3 The intersection between current deposition loading, historic loading, and sensitivity  
4 defines the ecological vulnerability to the effects of acidification. Freshwater aquatic and some  
5 terrestrial ecosystems, notably forests, are the ecosystem types which are most sensitive to  
6 acidification. The ISA reports that the principal factor governing the sensitivity of terrestrial and  
7 aquatic ecosystems to acidification from sulfur and nitrogen deposition is geology (particularly  
8 surficial geology). Geologic formations having low base cation supply generally underlie the  
9 watersheds of acid-sensitive lakes and streams. Other factors that contribute to the sensitivity of  
10 soils and surface waters to acidifying deposition include topography, soil chemistry, land use,  
11 and hydrologic flowpaths. Episodic and chronic acidification tends to occur in areas that have  
12 base-poor bedrock, high relief, and shallow soils (ISA 3.2.4.1).

13  
14 **2.1.3 What is the magnitude of ecosystem responses to acidifying deposition?**

15 Terrestrial and aquatic ecosystems differ in their response to acidifying deposition.  
16 Therefore the magnitude of ecosystem response is described separately for aquatic and terrestrial  
17 ecosystems in the following sections. The magnitude of response refers to both the severity of  
18 effects and the spatial extent of the U.S. which is affected.

19 **2.1.3.1 Aquatic**

20 Freshwater ecosystem surveys and monitoring in the eastern United States have been  
21 conducted by many programs since the mid-1980s, including EPA's Environmental Monitoring  
22 and Assessment Program (EMAP), National Surface Water Survey (NSWS), Temporally  
23 Integrated Monitoring of Ecosystems (TIME) (Stoddard, 1990), and Long-term Monitoring  
24 (LTM) (Ford et al., 1993; Stoddard et al., 1996) programs. Based on analyses of surface water  
25 data from these programs, New England, the Adirondack Mountains, the Appalachian Mountains  
26 (northern Appalachian Plateau and Ridge/Blue Ridge region), and the Upper Midwest contain  
27 the most sensitive lakes and streams (i.e., ANC less than about 50 µeq/L). Portions of northern  
28 Florida also contain many acidic and low-ANC lakes and streams, although the role of acidifying  
29 deposition in this region is less clear. The western U.S. contains many of the surface waters most  
30 sensitive to potential acidification effects, but with the exception of the Los Angeles Basin and  
31 surrounding areas, the levels of acidifying deposition are low in most areas. Therefore,

1 acidification of surface waters by acidic deposition is uncommon in the western U.S., and the  
2 extent of chronic surface water acidification that has occurred in that region to date has likely  
3 been very limited (**ISA 3.2.4.2 and REA 4.2.2**).

4 There are a number of species including fish, aquatic insects, other invertebrates and  
5 algae that are sensitive to acidification and cannot survive, compete, or reproduce in acidic  
6 waters (**ISA 3.2.3.3**). Decreases in ANC and pH have been shown to contribute to declines in  
7 species richness and declines in abundance of zooplankton, macroinvertebrates, and fish (Keller  
8 and Gunn 1995; Schindler et al., 1985). Reduced growth rates have been attributed to acid stress  
9 in a number of fish species including Atlantic salmon (*Salmo salar*), Chinook salmon  
10 (*Oncorhynchus tshawytscha*), lake trout (*Salvelinus namaycush*), rainbow trout (*Oncorhynchus*  
11 *mykiss*), brook trout (*Salvelinus Fontinalis*), and brown trout (*Salmo trutta*) (Baker et al., 1990).  
12 In response to small to moderate changes in acidity, acid-sensitive species are often replaced by  
13 other more acid-tolerant species, resulting in changes in community composition and richness.  
14 The effects of acidification are continuous, with more species being affected at higher degrees of  
15 acidification. At a point, typically a pH <4.5 and an ANC <0 µeq/L, complete to near-complete  
16 loss of many taxa of organisms occur, including fish and aquatic insect populations, whereas  
17 other taxa are reduced to only acidophilic species. These changes in taxa composition are  
18 associated with the high energy cost in maintaining physiological homeostasis, growth, and  
19 reproduction at low ANC levels (Schreck, 1981, 1982; Wedemeyer et al., 1990; **REA appendix**  
20 **2.3**). Decreases in species richness related to acidification have been observed in the Adirondack  
21 Mountains and Catskill Mountains of New York (Baker et al., 1996), New England and  
22 Pennsylvania (Haines and Baker, 1986), and Virginia (Bulger et al., 2000).

23 From the sensitive areas identified by the ISA, further “case study” analyses on aquatic  
24 ecosystems in the Adirondack Mountains and Shenandoah National Park were conducted to  
25 better characterize ecological risk associated with acidification (**REA Chapter 4**).

26 ANC is the most widely used indicator of acid sensitivity and has been found in various  
27 studies to be the best single indicator of the biological response and health of aquatic  
28 communities in acid-sensitive systems (Lien et al., 1992; Sullivan et al., 2006; ISA). In the  
29 REA, surface water trends in  $\text{SO}_4^{2-}$  and  $\text{NO}_3^-$  concentrations and ANC levels were analyzed to  
30 affirm the understanding that reductions in deposition could influence the risk of acidification.  
31 ANC values were categorized according to their effects on biota, as shown in Figure 2-2. [Need

1 to indicate the source for the categorization] Monitoring data from TIME/LTM and EMAP  
 2 programs were assessed for the years 1990 to 2006, and past, present, and future water quality  
 3 levels were estimated by both steady-state and dynamic biogeochemical models.

4

Category Label ANC Levels and Expected Ecological Effects		
Acute Concern	<0 µeq/L	Complete loss of fish populations is expected. Planktonic communities have extremely low diversity and are dominated by acidophilic taxa. The numbers of individuals in plankton species that are present are greatly reduced.
Severe Concern	0–20 µeq/L	Highly sensitive to episodic acidification. During episodes of high acidifying deposition, brook trout populations may experience lethal effects. The diversity and distribution of zooplankton communities decline sharply.
Elevated Concern	20–50 µeq/L	Fish species richness is greatly reduced (i.e., more than half of expected species can be missing). On average, brook trout populations experience sublethal effects, including loss of health, ability to reproduce, and fitness. Diversity and distribution of zooplankton communities decline.
Moderate Concern	50–100 µeq/L	Fish species richness begins to decline (i.e., sensitive species are lost from lakes). Brook trout populations are sensitive and variable, with possible sublethal effects. Diversity and distribution of zooplankton communities also begin to decline as species that are sensitive to acidifying deposition are affected.
Low Concern	>100 µeq/L	Fish species richness may be unaffected. Reproducing brook trout populations are expected where habitat is suitable. Zooplankton communities are unaffected and exhibit expected diversity and distribution.

5

6 **Table 2-1. Ecological effects associated with alternative levels of acid**  
 7 **neutralizing capacity (ANC)**

8

9 The most commonly used models of acidification are presented in Table 2-2. These models are  
 10 designed to be applied at the spatial scale of the watershed, with the exception of the SMART  
 11 model. Steady-state mass balance models, including the steady-state water chemistry (SSWC)  
 12 model are the most commonly used method for analysis of critical loads of acid deposition. The  
 13 steady-state models assume steady state conditions. The dynamic models consider how the  
 14 ecosystem may change through time. These models tend to require more data than the steady-  
 15 state models.

**Table 2-2 Summary several commonly used acidification models (See ISA Annex A for a more comprehensive list and discussion of acidification models)**

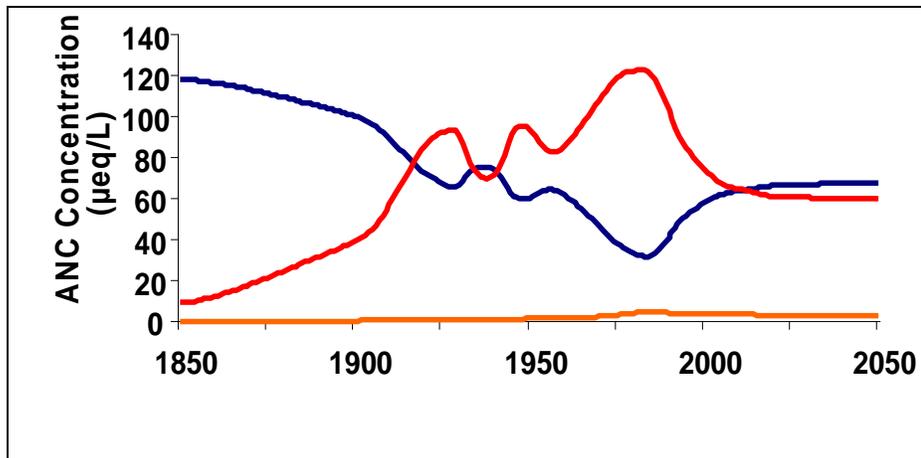
Model name	Dynamic or steady state	Model description
Steady-state mass balance models /Steady-state water Chemistry (SSWC)/	Steady-state	The basic principle is based on identifying the long-term average sources of acidity and alkalinity in order to determine the maximum acid input that will balance the system at a biogeochemical safe-limit. Several assumptions have been made in the steady state calculations. First, it is assumed that ion exchange is at steady state and there is no net change in basesaturation or no net transfer of ANC from soil solution to the ion exchange matrix. It is assumed that for N there is no net denitrification, adsorption or desorption and the N cycle is at steady state. Sulfate is also assumed to be at steady state: no sulfide oxidation, sulfate uptake, sulfate permanent fixation or sulfate reduction are significant. Simple hydrology is assumed where there is straight infiltration through the soil profile.
Model of Acidification of Groundwater in Catchment (MAGIC)	Dynamic	MAGIC is a lumped-parameter model of intermediate complexity, developed to predict the longterm effects of acidic deposition on surface water chemistry. The model simulates soil solution chemistry and surface water chemistry to predict the monthly and annual average concentrations of the major ions in these waters. MAGIC consists of: a section in which the concentrations of major ions are assumed to be governed by simultaneous reactions involving SO <sub>4</sub> <sup>2-</sup> adsorption, cation exchange, dissolution-precipitation- speciation of aluminum, and dissolution-speciation of inorganic carbon; and a mass balance section in which the flux of major ions to and from the soil is assumed to be controlled by atmospheric inputs, chemical weathering, net uptake and loss in biomass and losses to runoff.
PnET-BGC	Dynamic	PnET/BGC simulates major biogeochemical processes, such as forest canopy element transformations, hydrology, soil organic matter dynamics, N cycling, geochemical weathering, and chemical equilibrium reactions in solid and solution phases, and allows for simulations of land disturbance. The model uses mass transfer relationships to describe weathering, canopy interactions and surface water processes. Chemical equilibrium relationships describe anion adsorption, cation exchange and soil solution and surface water speciation. The model operates

<b>Table 2-2 Summary several commonly used acidification models (See ISA Annex A for a more comprehensive list and discussion of acidification models)</b>		
		on a monthly time step and is applied at the stand to small-watershed scale.
DayCent-Chem	Dynamic	DayCent-Chem links two widely accepted and tested models, one of daily biogeochemistry for forest, grassland, cropland, and savanna systems, DayCent (Parton et al., 1998), and the other of soil and water geochemical equilibrium, PHREEQC (Parkhurst and Appelo, 1999). The linked DayCent/PHREEQC model was created to capture the biogeochemical responses to atmospheric deposition and to explicitly consider those biogeochemical influences on soil and surface water chemistry. The linked model expands on DayCent's ability to simulate N, P, S, and C ecosystem dynamics by incorporating the reactions of many other chemical species in surface water.
Very Simple Dynamic (VSD) soil acidification model	Dynamic	This model is frequently used in Europe to simulate acidification effects in soils when observed data are sparse. The VSD model consists of a set of mass balance equations, describing the soil input-output relationships, and a set of equations describing the rate-limited and equilibrium soil processes. It only includes weathering, cation exchange, N immobilization processes, and a mass balance for cations, sulfur and N. In the VSD model, the various ecosystem processes have been limited to a few key processes. Processes that are not taken into account include canopy interactions; nutrient cycling processes; N fixation and NH <sub>4</sub> adsorption; SO <sub>4</sub> <sup>2-</sup> transformations (adsorption, uptake, immobilization, and reduction); formation and protonation of organic anions; and complexation of Al.
Simulation Model for Acidification's Regional Trends (SMART)	Dynamic	The the SMART model consists of a set of mass balance equations, describing soil input/output relationships, and a set of equations describing the rate-limited and equilibrium soil processes. It includes most of the assumptions and simplifications given for the VSD model. SMART models the exchange of Al, H, and divalent base cations using Gaines Thomas equations. Additionally, SO <sub>4</sub> <sup>2-</sup> adsorption is modeled using a Langmuir equation (as in MAGIC) and organic acids can be described as mono-, di-, or tri-protic. The SMART model has been developed with regional applications in mind, and an early example of an application to Europe can be found in De Vries et al. (1994).

1 The analyses of the Adirondack Case Study Area indicated that although wet deposition  
2 rates for SO<sub>2</sub> and NO<sub>x</sub> have been reduced since the mid-1990s, current concentrations are still  
3 well above simulated pre-acidification (1860) conditions. Modeling predicts NO<sub>3</sub><sup>-</sup> and SO<sub>4</sub><sup>2-</sup> are  
4 17- and 5-fold higher, respectively, in 2006 than under simulated pre-acidification conditions.  
5 Based on the 2006 Model of Acidification of Groundwater in Catchment (MAGIC) simulations,  
6 the estimated average ANC across the 44 lakes in the Adirondack Case Study Area is 62.1 µeq/L  
7 (± 15.7 µeq/L); 78 % of all monitored lakes in the Adirondack Case Study Area have a current  
8 risk of *Elevated*, *Severe*, or *Acute*. Of the 78%, 18% are chronically acidic (REA 4.2.4.2).

9 Based on a N and S deposition scenario that maintains current emission levels to 2020  
10 and 2050, the simulation forecast indicates there would be no improvement in water quality in  
11 the Adirondack Case Study Area. The percentage of lakes within the *Elevated to Acute Concern*  
12 classes remains the same from 2020 to 2050.

13

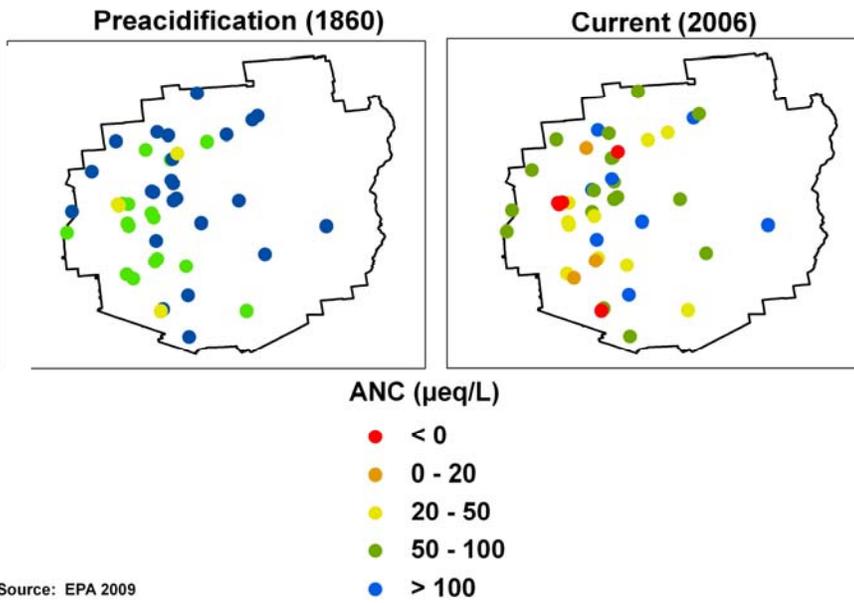


14

15 **Figure 2-2.** Average NO<sub>3</sub><sup>-</sup> concentrations (orange), SO<sub>4</sub><sup>2-</sup> concentrations  
16 (red), and ANC (blue) across the 44 lakes in the Adirondack  
17 Case Study Area modeled using MAGIC for the period 1850  
18 to 2050.

19

ANC Preacidification (1860) and Current Condition (2006)

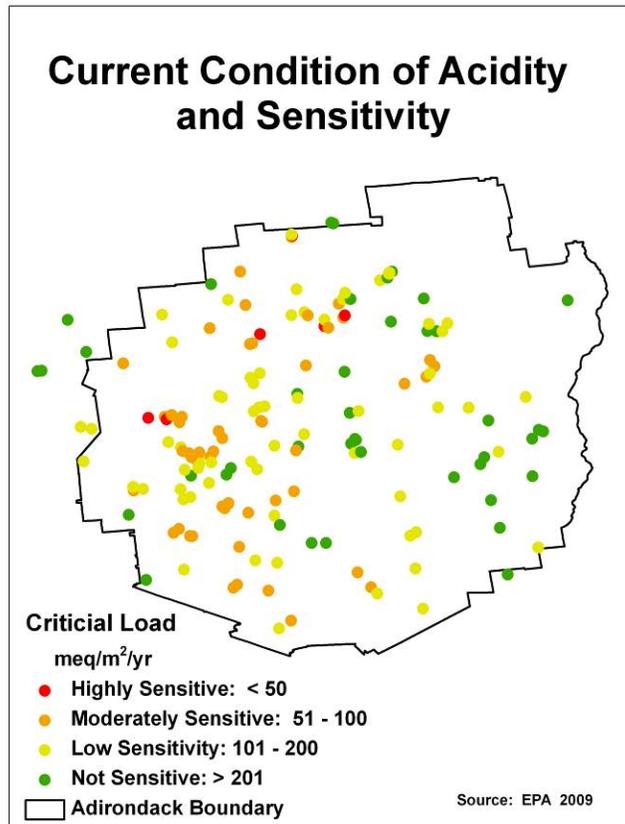


Source: EPA 2009

1

2 **Figure 2-3. ANC concentrations of preacidification (1860) and 2006 conditions**  
3 **based on hindcasts of 44 lakes in the Adirondack Case Study Area**  
4 **modeled using MAGIC.**

5



1

2 **Figure 2-4. Critical loads of acidifying deposition that each surface water location**  
 3 **can receive in the Adirondack Case Study Area while maintaining or**  
 4 **exceeding an ANC concentration of 50 µeq/L based on 2002 data.**  
 5 **Watersheds with critical load values <100 meq/m<sup>2</sup>/yr (red and orange**  
 6 **circles) are most sensitive to surface water acidification, whereas**  
 7 **watersheds with values >100 meq/m<sup>2</sup>/yr (yellow and green circles) are**  
 8 **less sensitive sites.**

9

10 Note that studies on fish species richness in the Adirondacks Case Study Area  
 11 demonstrated the effect of acidification. Of the 53 fish species recorded in Adirondack Case  
 12 Study Area lakes, only 27 species were found in lakes with a pH <6.0. The 26 species missing  
 13 from lakes with a pH <6.0 include important recreational species, such as Atlantic salmon, tiger  
 14 trout (*Salmo trutta X Salvelinus fontinalis*), redbreast sunfish (*Lepomis auritus*), bluegill  
 15 (*Lepomis macrochirus*), tiger musky (*Esox masquinongy X lucius*), walleye (*Sander vitreus*),  
 16 alewife (*Alosa pseudoharengus*), and kokanee (*Oncorhynchus nerka*) (Kretser et al., 1989), as  
 17 well as ecologically important minnows that are commonly consumed by sport fish. A survey of  
 18 1,469 lakes in the late 1980s found 346 lakes to be devoid of fish. Among lakes with fish, there

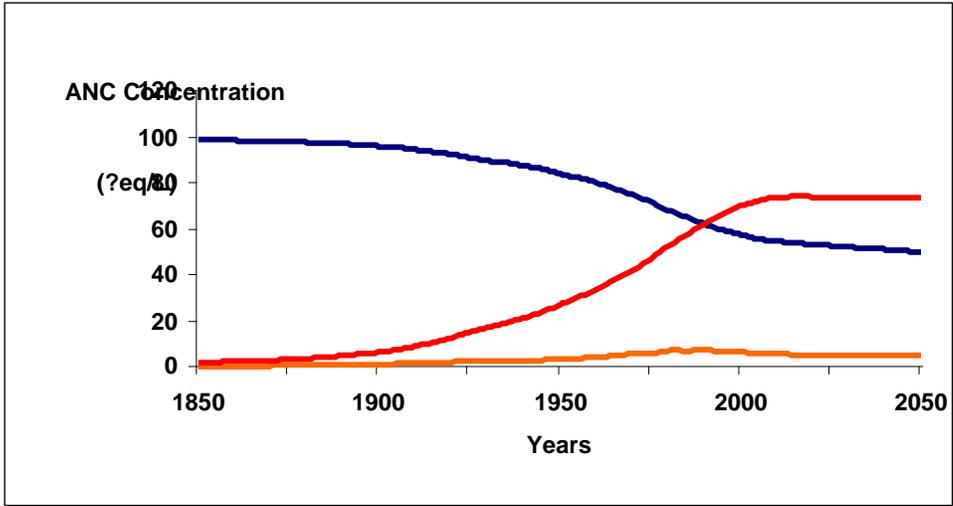
1 was a relationship between the number of fish species and lake pH, ranging from about one  
2 species per lake for lakes having a pH <4.5 to about six species per lake for lakes having a pH  
3 >6.5 (Driscoll et al., 2001; Kretser et al., 1989). In the Adirondacks, a positive relationship  
4 exists between the pH and ANC in lakes and the number of fish species present in those lakes  
5 **(ISA 3.2.3.4)**.

6 Since the mid-1990s, streams in the Shenandoah Case Study Area have shown slight  
7 declines in NO<sub>3</sub><sup>-</sup> and SO<sub>4</sub><sup>2-</sup> concentrations in surface waters. The 2006 concentrations are still  
8 above pre-acidification (1860) conditions. MAGIC modeling predicts surface water  
9 concentrations of NO<sub>3</sub><sup>-</sup> and SO<sub>4</sub><sup>2-</sup> are 10- and 32-fold higher, respectively, in 2006 than in 1860.  
10 The estimated average ANC across 60 streams in the Shenandoah Case Study Area is 57.9 µeq/L  
11 (± 4.5 µeq/L). 55% of all monitored streams in the Shenandoah Case Study Area have a current  
12 risk of *Elevated*, *Severe*, or *Acute*. Of the 55%, 18% are chronically acidic today **(REA 4.2.4.3)**

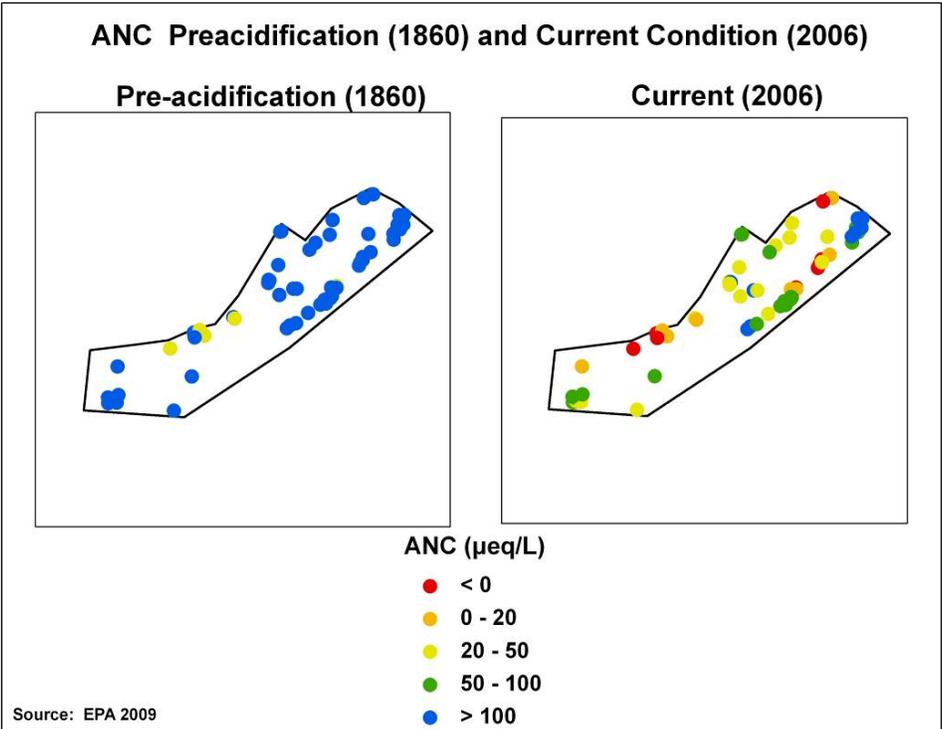
13 Based on a deposition scenario for this study area that maintains current emission levels  
14 from 2020 to 2050, the simulation forecast indicates that a large number of streams still have  
15 *Elevated* to *Acute* problems with acidity. In fact, from 2006 to 2050, the percentage of streams  
16 with *Acute Concern* are predicted to increase by 5%, while the percentage of streams in  
17 *Moderate Concern* decreases by 5%.

18 Biological effects of increased acidification documented in the Shenandoah Case Study  
19 Area include a decrease in the condition factor in blacknose dace (Dennis and Bulgar 1995,  
20 Bulgar et al., 1999) and a decrease in fish biodiversity associated with decreasing stream ANC  
21 (Bulger et al., 1995; Dennis and Bulger, 1995; Dennis et al., 1995; MacAvoy and Bulger, 1995,  
22 Bulgar et al., 1999). On average, the fish species richness is lower by one fish species for every  
23 21 µeq/L decrease in ANC in Shenandoah National Park streams **(ISA 3.2.3.4)**.

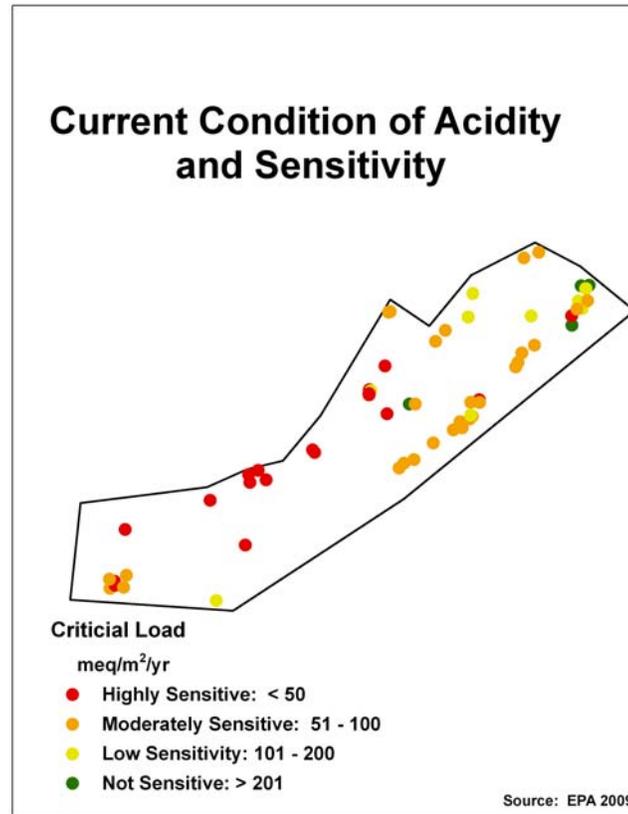
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**Figure 2-5.** Average  $\text{NO}_3^-$  concentrations (orange),  $\text{SO}_4^{2-}$  concentrations (red), and ANC (blue) levels for the 60 streams in the Shenandoah Case Study Area modeled using MAGIC for the period 1850 to 2050.



**Figure 2-6.** ANC levels of 1860 (preacidification) and 2006 (current) conditions based on hindcasts of 60 streams in the Shenandoah Case Study Area modeled using MAGIC.



2

3 **Figure 2-7. Critical loads of surface water acidity for an ANC of 50 µeq/L for**  
 4 **Shenandoah Case Study Area streams. Each dot represents an**  
 5 **estimated amount of acidifying deposition (i.e., critical load) that each**  
 6 **stream's watershed can receive and still maintain a surface water**  
 7 **ANC >50 µeq/L. Watersheds with critical load values <100 meq/m<sup>2</sup>/yr**  
 8 **(red and orange circles) are most sensitive to surface water**  
 9 **acidification, whereas watersheds with values >100 meq/m<sup>2</sup>/yr (yellow**  
 10 **and green circles) are less sensitive sites.**

11

### 12 2.1.3.2 Terrestrial Acidification

13 The ISA identified a variety of indicators that can be used to measure the effects of  
 14 acidification in soils. Most effects of terrestrial acidification are observed in sensitive forest  
 15 ecosystem in the U.S. Tree health has been linked to the availability of base cations (Bc) in soil  
 16 (such as Ca<sup>2+</sup>, Mg<sup>2+</sup> and potassium), as well as soil Al content. Tree species show a range of  
 17 sensitivities to Ca/Al and Bc/Al soil molar ratios, therefore these are good chemical indicators  
 18 because they directly relate to the biological effects. Critical Bc/Al molar ratios for a large

1 variety of tree species ranged from 0.2 to 0.8 (Sverdrup and Warfvinge, 1993, a meta-data  
2 analysis of laboratory and field studies). This range is similar to critical ratios of Ca/Al. Plant  
3 toxicity or nutrient antagonism was reported to occur at Ca/Al molar ratios ranging from 0.2 to  
4 2.5 (Cronan and Grigal, 1995; meta-data assessment) (**REA pg 4-54, REA Appendix 5**).

5         There has been no systematic national survey of terrestrial ecosystems to determine the  
6 extent and distribution of terrestrial ecosystem sensitivity to the effects of acidifying deposition.  
7 However, one preliminary national evaluation estimated that ~15% of forest ecosystems in the  
8 U.S. exceed the estimated critical load based on soil ANC leaching for S and N deposition by  
9 >250 eq ha<sup>-1</sup> yr<sup>-1</sup> (McNulty et al., 2007). Forests of the Adirondack Mountains of New York,  
10 Green Mountains of Vermont, White Mountains of New Hampshire, the Allegheny Plateau of  
11 Pennsylvania, and high-elevation forest ecosystems in the southern Appalachians are the regions  
12 most sensitive to terrestrial acidification effects from acidifying deposition (**ISA 3.2.4.2**). While  
13 studies show some recovery of surface waters, there are widespread measurements of ongoing  
14 depletion of exchangeable base cations in forest soils in the northeastern U.S. despite recent  
15 decreases in acidifying deposition, indicating a slow recovery time.

16         In the REA, a critical load analysis was performed for sugar maple and red spruce forests  
17 in the eastern United States by using Bc/Al ratio in acidified forest soils as an indicator to assess  
18 the impact of nitrogen and sulfur deposition on tree health. These are the two most commonly  
19 studied tree species in North America for effects of acidification. At a Bc/Al ratio of 1.2, red  
20 spruce growth can be decreased by 20%. Sugar maple growth can be decreased by 20% at a  
21 Bc/Al ratio of 0.6 (**REA 4.4**). The REA analysis determined the health of at least a portion of the  
22 sugar maple and red spruce growing in the United States may have been compromised with  
23 acidifying total nitrogen and sulfur deposition. Specifically, total nitrogen and sulfur deposition  
24 levels exceeded three selected critical loads for tree growth in 3% to 75% of all sugar maple  
25 plots across 24 states. For red spruce, total nitrogen and sulfur deposition levels exceeded three  
26 selected critical loads in 3% to 36% of all red spruce plots across eight states (**REA 4.4**).

#### 27 28 **2.1.4 What are the key uncertainties associated with acidification?**

29         There are different levels of uncertainty associated with relationships between deposition,  
30 ecological effects and ecological indicators. In Chapter 7 of the REA, the case study analyses  
31 associated with each targeted effect area were synthesized by identifying the strengths,

1 limitations, and uncertainties associated with the available data, modeling approach, and  
2 relationship between the selected ecological indicator and atmospheric deposition as described  
3 by the ecological effect function (Figure 1-1). The key uncertainties were characterized as  
4 follows to evaluate the strength of the scientific basis for setting a national standard to protect  
5 against a given effect (**REA 7.0**):

- 6 • **Data Availability: *high, medium or low quality***. This criterion is based on the  
7 availability and robustness of data sets, monitoring networks, availability of data that  
8 allows for extrapolation to larger assessment areas, and input parameters for modeling  
9 and developing the ecological effect function. The scientific basis for the ecological  
10 indicator selected is also incorporated into this criterion.
- 11 • **Modeling Approach: *high, fairly high, intermediate, or low confidence***. This value is  
12 based on the strengths and limitations of the models used in the analysis and how  
13 accepted they are by the scientific community for their application in this analysis.
- 14 • **Ecological Effect Function: *high, fairly high, intermediate, or low confidence***. This  
15 ranking is based on how well the ecological effect function describes the relationship  
16 between atmospheric deposition and the ecological indicator of an effect.

#### 17 18 **2.1.4.1 Aquatic Acidification**

19 The REA concludes that the available data are robust and considered *high quality*. There  
20 is high confidence about the use of these data and their value for extrapolating to a larger  
21 regional population of lakes. The EPA TIME/LTM network represents a source of long-term,  
22 representative sampling. Data on sulfate concentrations, nitrate concentrations and ANC from  
23 1990 to 2006 used for this analysis as well as EPA EMAP and REMAP surveys, provide  
24 considerable data on surface water trends.

25 There is *fairly high confidence* associated with modeling and input parameters.  
26 Uncertainties in water quality estimates (.i.e., ANC) from MAGIC was derived from multiple  
27 site calibrations. The 95% confidence interval for pre-acidification of lakes was an average of 15  
28  $\mu\text{eq/L}$  difference in ANC concentrations or 10% and 8  $\mu\text{eq/L}$  or 5% for streams (**REA 7.1.2**).  
29 The use of the critical load model used to estimate aquatic critical loads is limited by the  
30 uncertainties associated with runoff and surface water measurements and in estimating the  
31 catchment supply of base cations from the weathering of bedrock and soils (McNulty et al.,

1 2007). To propagate uncertainty in the model parameters, Monte Carlo methods were employed  
2 to develop an inverse function of exceedences. There is *high confidence* associated with the  
3 ecological effect function developed for aquatic acidification. In calculating the ANC function,  
4 the depositional load for N or S is fixed by the deposition of the other, so deposition for either  
5 will never be zero (**Figure 7.1-6 REA**).

#### 6 7 **2.1.4.2 Terrestrial Acidification**

8 The available data used to quantify the targeted effect of terrestrial acidification are  
9 robust and considered *high quality*. The USFS-Kane Experimental Forest and significant  
10 amounts of research work in the Allegheny Plateau have produced extensive, peer-reviewed data  
11 sets. A meta-analysis of laboratory studies showed that tree growth was decreased by 20%  
12 relative to controls for BC/Al ratios (**ISA 7.2.1 and Figure 7.2-1**). Sugar maple and red spruce  
13 were the focus of the REA since they are demonstrated to be negatively affected by soil available  
14  $\text{Ca}^{2+}$  depletion and high concentrations of available Al, and occur in areas that receive high  
15 acidifying deposition, There is high confidence about the use of the REA terrestrial acidification  
16 data and their value for extrapolating to a larger regional population of forests.

17 There is *high confidence* associated with the models, input parameters, and assessment of  
18 uncertainty used in the case study for terrestrial acidification. The Simple Mass Balance (SMB)  
19 model, a commonly used and widely applied approach for estimating critical loads, was used in  
20 the REA analysis (**ISA 7.2.2**). There is fairly high confidence associated with the ecological  
21 effect function developed for terrestrial acidification (**REA 7.2.3**).

## 22 23 **2.2 NITROGEN ENRICHMENT: EVIDENCE OF EFFECTS ON STRUCTURE AND** 24 **FUNCTION OF TERRESTRIAL AND FRESHWATER ECOSYSTEMS**

25 The following summary is a concise overview of the known or anticipated effects caused  
26 by nitrogen nutrient enrichment to ecosystems within the United States. Nutrient-enrichment  
27 affects terrestrial, freshwater and estuarine ecosystems. Nitrogen deposition is a major source of  
28 anthropogenic nitrogen. For many terrestrial and freshwater ecosystems other sources of  
29 nitrogen including fertilizer and waste treatment are greater than deposition. Nitrogen deposition  
30 often contributes to nitrogen-enrichment effects in estuaries, but does not drive the effects since  
31 other sources of N greatly exceed N deposition. Both oxides of nitrogen and reduced forms of

1 nitrogen, (e.g. NH<sub>x</sub>) contribute to nitrogen deposition. For the most part, nitrogen effects on  
2 ecosystems do not depend on whether the nitrogen is in oxidized or reduced form. Thus, this  
3 summary focuses on the effects of nitrogen deposition in total. We address the issue of  
4 incorporating the relative contributions of oxidized and reduced nitrogen into the standards in  
5 Chapters 5 and 8.

6

### 7 **2.2.1 What is the nature of terrestrial and freshwater ecosystem responses to reactive** 8 **nitrogen and/ sulfur deposition?**

9 The ISA found that deposition of nitrogen, including NO<sub>x</sub> and NH<sub>x</sub>, leads to the nitrogen  
10 enrichment of ecosystems (EPA 2008). In the process of nitrogen enrichment, biogeochemical  
11 components of terrestrial and freshwater aquatic ecosystems are altered in a way that leads to  
12 effects on biological organisms.

13 *The evidence is sufficient to infer a **causal** relationship between N deposition and the*  
14 *alteration of biogeochemical cycling in terrestrial ecosystems (ISA 4.3.1.1 and 3.3.2.1).* This is  
15 supported by numerous observational, deposition gradient and field addition experiments in  
16 sensitive ecosystems. Stoddard (1994) identified the leaching of NO<sub>3</sub><sup>-</sup> in soil drainage waters and  
17 the export of NO<sub>3</sub><sup>-</sup> in stream water as two of the primary indicators of N enrichment. Several N-  
18 addition studies indicate that NO<sub>3</sub><sup>-</sup> leaching is induced by chronic additions of N (Edwards et al.,  
19 2002b; Kahl et al., 1999; Peterjohn et al., 1996; Norton et al., 1999). Aber et al. (2003) found  
20 that surface water NO<sub>3</sub><sup>-</sup> concentrations exceeded 1 µeq/L in watersheds receiving about 9 to 13  
21 kg N/ha/yr of atmospheric N deposition. N deposition disrupts the nutrient balance of  
22 ecosystems with numerous biogeochemical effects. The chemical indicators that are typically  
23 measured include NO<sub>3</sub><sup>-</sup> leaching, soil C:N ratio, rates of N mineralization, nitrification,  
24 denitrification, foliar N concentration, and soil water NO<sub>3</sub><sup>-</sup> and NH<sub>4</sub><sup>+</sup> concentrations. Note that  
25 N saturation (N leaching from ecosystems) does not need to occur to cause effects. Substantial  
26 leaching of NO<sub>3</sub><sup>-</sup> from forest soils to stream water can acidify downstream waters, leading to  
27 effects described in the previous section on aquatic acidification. Due to the complexity of  
28 interactions between the N and C cycling, the effects of N on C budgets (quantified input and  
29 output of C to the ecosystem) are variable. Regional trends in net ecosystem productivity (NEP)  
30 of forests (not managed for silviculture) have been estimated through models based on gradient  
31 studies and meta-analysis. Atmospheric N deposition has been shown to cause increased litter

1 accumulation and carbon storage in above-ground woody biomass. In the West, this has lead to  
2 increased susceptibility to more severe fires. Less is known regarding the effects of N deposition  
3 on C budgets of non-forest ecosystems.

4 *The evidence is sufficient to infer a **causal** relationship between N deposition on the*  
5 *alteration of species richness, species composition and biodiversity in terrestrial ecosystems*  
6 **(ISA 4.3.1.2)**. Some organisms and ecosystems are more sensitive to N deposition and effects of  
7 N deposition are not observed in all habitats. The most sensitive terrestrial taxa to N deposition  
8 are lichens. Empirical evidence indicates that lichens in the U.S. are affected by deposition levels  
9 as low as 3 kg N/ha/yr. Alpine ecosystems are also sensitive to N deposition, changes in an  
10 individual species (*Carex rupestris*) were estimated to occur at deposition levels near 4 kg N  
11 /ha/yr and modeling indicates that deposition levels near 10 kg N/ha/yr alter plant community  
12 assemblages. In several grassland ecosystems, reduced species diversity and an increase in non-  
13 native, invasive species are associated with N deposition (Clark and Tillman, 2008; Schwinning  
14 et al., 2005).

15 *In freshwater ecosystems, the evidence is sufficient to infer a **causal** relationship between*  
16 *N deposition and the alteration of biogeochemical cycling in freshwater aquatic ecosystems (ISA*  
17 **3.3.2.3)**. N deposition is the main source of N enrichment to headwater streams, lower order  
18 streams and high elevation lakes. The most common chemical indicators that were studied  
19 included  $\text{NO}_3^-$  and dissolved inorganic nitrogen (DIN) concentration in surface waters as well as  
20 Chl a:total P ratio. Elevated surface water  $\text{NO}_3^-$  concentrations occur in both the eastern and  
21 western U.S. Bergstrom and Jansson (2006) report a significant correlation between N deposition  
22 and lake biogeochemistry by identifying a correlation between wet deposition and [DIN] and Chl  
23 a: Total P. Recent evidence provides examples of lakes and streams that are limited by N and  
24 show signs of eutrophication in response to N addition.

25 *The evidence is sufficient to infer a **causal** relationship between N deposition and the*  
26 *alteration of species richness, species composition and biodiversity in freshwater aquatic*  
27 *ecosystems (ISA 3.3.5.3)*. Increased N deposition can cause a shift in community composition  
28 and reduce algal biodiversity, especially in sensitive oligotrophic lakes.

1 **2.2.2 What types of ecosystems are sensitive to such effects? How are these responses**  
2 **affected by atmospheric, ecological, and landscape factors?**

3 The numerous ecosystem types that occur across the U.S. have a broad range of  
4 sensitivity to N deposition (Clark and Tilman 2008; Aber et al., 2003; Fenn et al., 2003; Rueth et  
5 al., 2003; Egerton-Warburton and Allen 2000; Williams et al., 1996; and additional studies  
6 summarized in **Table 4-4 ISA**). Increased deposition to N-limited ecosystems can lead to  
7 production increases that may be either beneficial or adverse depending on the system and  
8 management goals.

9 Organisms in their natural environment are commonly adapted to a specific regime of  
10 nutrient availability. Change in the availability of one important nutrient, such as N, may result  
11 in imbalance in ecological stoichiometry, with effects on ecosystem processes, structure and  
12 function (Sterner and Elser, 2002). In general, N deposition to terrestrial ecosystems causes  
13 accelerated growth rates in some species deemed desirable in commercial forests but may lead to  
14 altered competitive interactions among species and nutrient imbalances, ultimately affecting  
15 biodiversity. The onset of these effects occurs with N deposition levels as low as 3 kg N/ha/yr in  
16 sensitive terrestrial ecosystems to N deposition. In aquatic ecosystems, N that is both leached  
17 from the soil and directly deposited to the water surface can pollute the surface water. This  
18 causes alteration of the diatom community at levels as low as 1.5 kg N/ha/yr in sensitive  
19 freshwater ecosystems.

20 The degree of ecosystem effects lies at the intersection of N loading and N-sensitivity.  
21 N-sensitivity is predominately driven by the degree to which growth is limited by nitrogen  
22 availability. Grasslands in the western United States are typically N-limited ecosystems  
23 dominated by a diverse mix of perennial forbs and grass species (Clark and Tilman, 2008;  
24 Suding et al., 2005). A meta-analysis by LeBauer and Treseder (2008) indicated that N  
25 fertilization increased aboveground growth in all non-forest ecosystems except for deserts. In  
26 other words, almost all terrestrial ecosystems are N-limited and will be altered by the addition of  
27 anthropogenic nitrogen (LeBauer and Treseder, 2008). Likewise, a freshwater lake or stream  
28 must be N-limited to be sensitive to N-mediated eutrophication. There are many examples of  
29 fresh waters that are N-limited or N and phosphorous (P) co-limited (**ISA 3.3.3.2**). In a meta-  
30 analysis that included 653 datasets, Elser et al. (2007) found that N-limitation occurred as  
31 frequently as P-limitation in freshwater ecosystems. Additional factors that govern the sensitivity

1 of ecosystems to nutrient enrichment from N deposition include rates and form of N deposition,  
2 elevation, climate, species composition, plant growth rate, length of growing season, and soil N  
3 retention capacity. (ISA 4.3). Less is known about the extent and distribution of the terrestrial  
4 ecosystems in the U.S. that are most sensitive to the effects of nutrient enrichment from  
5 atmospheric N deposition compared to acidification.

## 6 7 **2.2.3 What is the magnitude of ecosystem responses to nitrogen deposition?**

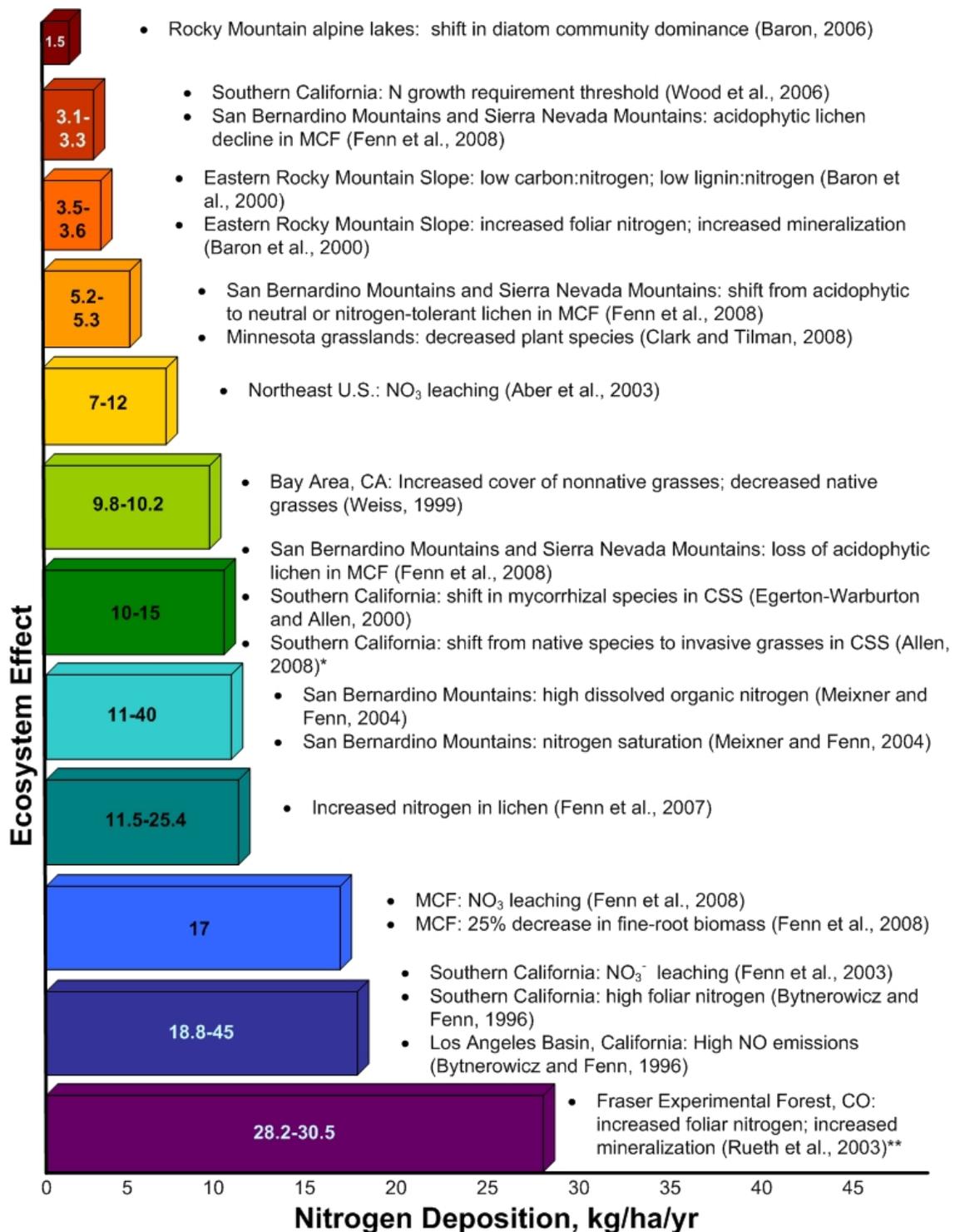
### 8 **2.2.3.1 Terrestrial**

9 Little is known about the full extent and distribution of the terrestrial ecosystems in the  
10 U.S. that are most sensitive to impacts caused by nutrient enrichment from atmospheric N  
11 deposition. As previously stated, most terrestrial ecosystems are N-limited, therefore they are  
12 sensitive to perturbation caused by N additions (LeBauer and Treseder, 2008). Effects are most  
13 likely to occur where areas of relatively high atmospheric N deposition intersect with N-limited  
14 plant communities. The alpine ecosystems of the Colorado Front Range, chaparral watersheds of  
15 the Sierra Nevada, lichen and vascular plant communities in the San Bernardino Mountains and  
16 the Pacific Northwest, and the southern California coastal sage scrub (CSS) community are  
17 among the most sensitive terrestrial ecosystems. There is growing evidence that existing  
18 grassland ecosystems in the western United States are being altered by elevated levels of N  
19 inputs, including inputs from atmospheric deposition (Clark and Tilman, 2008; Suding et al.,  
20 2005).

21 In the eastern U.S., the degree of N saturation of the terrestrial ecosystem is often  
22 assessed in terms of the degree of  $\text{NO}_3^-$  leaching from watershed soils into ground water or  
23 surface water. Stoddard (1994) estimated the number of surface waters at different stages of  
24 saturation across several regions in the eastern U.S. Of the 85 northeastern watersheds examined  
25 60% were in Stage 1 or Stage 2 of N saturation on a scale of 0 (background or pretreatment) to 3  
26 (visible decline). Of the northeastern sites for which adequate data were available for assessment,  
27 those in Stage 1 or 2 were most prevalent in the Adirondack and Catskill Mountains. Effects on  
28 individual plant species have not been well studied in the U.S. More is known about the  
29 sensitivity of particular plant communities. Based largely on results obtained in more extensive  
30 studies conducted in Europe, it is expected that the more sensitive terrestrial ecosystems include

1 hardwood forests, alpine meadows, arid and semi-arid lands, and grassland ecosystems (**ISA**  
2 **3.8.2**).

3         The REA used published research results (**REA 5.3.1 and ISA Table 4.4**) to identify  
4 meaningful ecological benchmarks associated with different levels of atmospheric nitrogen  
5 deposition. These are given by Figure 2-8. The sensitive areas and ecological indicators  
6 identified by the ISA were analyzed further in the REA to create a national map that illustrates  
7 effects observed from ambient and experimental atmospheric nitrogen deposition loads in  
8 relation to Community Multi-scale Air Quality (CMAQ) 2002 modeling results and NADP  
9 monitoring data. This map, reproduced in Figure 2-9, depicts the sites where empirical effects of  
10 terrestrial nutrient enrichment have been observed and site proximity to elevated atmospheric N  
11 deposition.

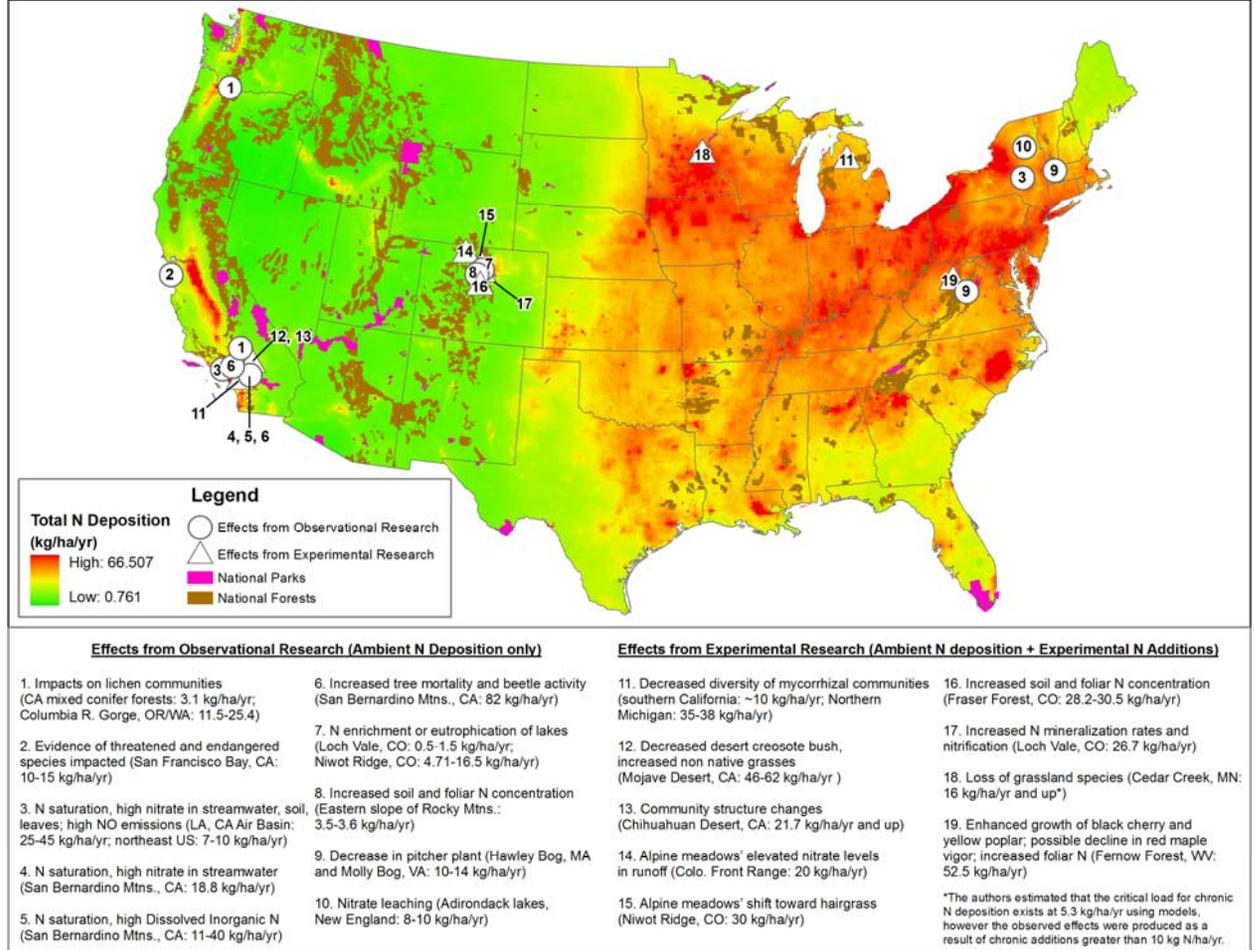


\* Personal communication, 2008. Also referenced in Bobbink et. al., 2010, Ecological Applications, 20(1):30-59 and USDS FS, 2010, [http://www.nrs.fs.fed.us/clean\\_air\\_water/clean\\_water/critical\\_loads/local-resources/docs/Empirical\\_CLS\\_of\\_N\\_100414.pdf](http://www.nrs.fs.fed.us/clean_air_water/clean_water/critical_loads/local-resources/docs/Empirical_CLS_of_N_100414.pdf)

\*\*Nitrogen deposition levels include ambient and experimental additions.

1  
2

**Figure 2-8. Benchmarks of atmospheric nitrogen deposition for several ecosystem indicators (REA 5.3.1.2) MCF-Mixed Conifer Forest, CSS-Coastal Sage Scrub**



1

2 **Figure 2-9 Observed effects from ambient and experimental atmospheric nitrogen deposition loads in relation to**  
 3 **using CMAQ 2002 modeling results and NADP monitoring data.** Citations for effect results are from the ISA, Table  
 4 4.4 (U.S. EPA, 2008) 1=Fenn et. al. (2008), 2=Weiss (1999), 3=Bytnerowicz and Fenn (1996), 4=Fenn et al. (2000), 5= Meixner and  
 5 Fenn (2004), 6=Jones et al. 2004, 7=Baron (2006), 8=Baron et al. (2000), 9=Gotelli and Ellison (2002), 10=Stoddard et al. (1994),  
 6 11=Egerton Warburton and Allen (2000), 12=Brooks (2003), 13=Baez et al. (2007), 14=Bowman et al. (2006), 15=Bowman et al.  
 7 (1995), 16=Rueth et al. (2003), 17=DeWalle et al. (2006), 18=Clark and Tillman (2008), 19=Rueth et al. 2003

1 Based on information in the ISA and initial analysis in the REA, further case  
2 study analyses on terrestrial nutrient enrichment of ecosystems were developed for the  
3 CCS community and Mixed Conifer Forest (MCF) (EPA 2009). Geographic information  
4 systems (GIS) analysis supported a qualitative review of past field research to identify  
5 ecological benchmarks associated with CSS and mycorrhizal communities, as well as  
6 MCF's nutrient-sensitive acidophyte lichen communities, fine-root biomass in Ponderosa  
7 pine, and leached nitrate in receiving waters.

8 The ecological benchmarks that were identified for the CSS and the MCF are  
9 included in the suite of benchmarks identified in the ISA (**ISA 3.3**). There are sufficient  
10 data to confidently relate the ecological effect to a loading of atmospheric nitrogen. For  
11 the CSS community, the following ecological benchmarks were identified:

- 12 • 3.3 kg N/ha/yr – the amount of nitrogen uptake by a vigorous stand of CSS; above  
13 this level, nitrogen may no longer be limiting
- 14 • 10 kg N/ha/yr – mycorrhizal community changes

15 For the MCF community, the following ecological benchmarks were identified:

- 16 • 3.1 kg N/ha/yr – shift from sensitive to tolerant lichen species
- 17 • 5.2 kg N/ha/yr – dominance of the tolerant lichen species
- 18 • 10.2 kg N/ha/yr – loss of sensitive lichen species
- 19 • 17 kg N/ha/yr – leaching of nitrate into streams.

20  
21 These benchmarks, ranging from 3.1 to 17 kg N/ha/yr, were compared to 2002  
22 CMAQ/NADP data to discern any associations between atmospheric deposition and  
23 changing communities. Evidence supports the finding that nitrogen alters CSS and MCF.  
24 Key findings include the following: 2002 CMAQ/NADP nitrogen deposition data show  
25 that the 3.3 kg N/ha/yr benchmark has been exceeded in more than 93% of CSS areas  
26 (654,048 ha). These deposition levels are a driving force in the degradation of CSS  
27 communities. Although CSS decline has been observed in the absence of fire, the  
28 contributions of deposition and fire to the CSS decline require further research. CSS is  
29 fragmented into many small parcels, and the 2002 CMAQ/NADP 12-km grid data are not  
30 fine enough to fully validate the relationship between CSS distribution, nitrogen  
31 deposition, and fire. 2002 CMAQ/NADP nitrogen deposition data exceeds the 3.1 kg

1 N/ha/yr benchmark in more than 38% (1,099,133 ha) of MCF areas, and nitrate leaching  
2 has been observed in surface waters. Ozone effects confound nitrogen effects on MCF  
3 acidophyte lichen, and the interrelationship between fire and nitrogen cycling requires  
4 additional research.

### 6 **2.2.3.2 Freshwater**

7 The magnitude of ecosystem response may be thought of on two time scales,  
8 current conditions and how ecosystems have been altered since the onset of  
9 anthropogenic N deposition. As noted previously, Elser et al. (2008) found that N-  
10 limitation occurs as frequently as P-limitation in freshwater ecosystems (**ISA 3.3.3.2**).  
11 Recently, a comprehensive study of available data from the northern hemisphere surveys  
12 of lakes along gradients of N deposition show increased inorganic N concentration and  
13 productivity to be correlated with atmospheric N deposition (Bergström and Jansson  
14 2006). The results are unequivocal evidence of N limitation in lakes with low ambient  
15 inputs of N, and increased N concentrations in lakes receiving N solely from atmospheric  
16 N deposition (Bergström and Jansson, 2006). These authors suggested that most lakes in  
17 the northern hemisphere may have originally been N-limited, and that atmospheric N  
18 deposition has changed the balance of N and P in lakes.

19 Available data suggest that the increases in total N deposition do not have to be  
20 large to elicit an ecological effect. For example, a hindcasting exercise determined that  
21 the change in Rocky Mountain National Park lake algae that occurred between 1850 and  
22 1964 was associated with an increase in wet N deposition that was only about 1.5 kg  
23 N/ha (Baron, 2006). Similar changes inferred from lake sediment cores of the Beartooth  
24 Mountains of Wyoming also occurred at about 1.5 kg N/ha deposition (Saros et al.,  
25 2003). Pre-industrial inorganic N deposition is estimated to have been only 0.1 to 0.7 kg  
26 N/ha based on measurements from remote parts of the world (Galloway et al., 1995;  
27 Holland et al., 1999). In the western U.S., pre-industrial, or background, inorganic N  
28 deposition was estimated by (Holland et al., 1999) to range from 0.4 to 0.7 kg N/ha/yr.

29 Eutrophication effects from N deposition are most likely to be manifested in  
30 undisturbed, low nutrient surface waters such as those found in the higher elevation areas  
31 of the western U.S. The most severe eutrophication from N deposition effects is expected

1 downwind of major urban and agricultural centers. High concentrations of lake or  
2 streamwater  $\text{NO}_3^-$ , indicative of ecosystem saturation, have been found at a variety of  
3 locations throughout the U.S., including the San Bernardino and San Gabriel Mountains  
4 within the Los Angeles Air Basin (Fenn et al., 1996), the Front Range of Colorado  
5 (Baron et al., 1994; Williams et al., 1996), the Allegheny mountains of West Virginia  
6 (Gilliam et al., 1996), the Catskill Mountains of New York (Murdoch and Stoddard,  
7 1992; Stoddard, 1994), the Adirondack Mountains of New York (Wigington et al., 1996),  
8 and the Great Smoky Mountains in Tennessee (Cook et al., 1994) (**ISA 3.3.8**).

### 10 **2.2.3.3 Estuaries**

11 In contrast to terrestrial and freshwater systems, atmospheric N load to estuaries  
12 contributes to the total load but does not necessarily drive the effects since other sources  
13 of N greatly exceed N deposition. In estuaries, N-loading from multiple anthropogenic  
14 and non-anthropogenic pathways leads to water quality deterioration, resulting in  
15 numerous effects including hypoxic zones, species mortality, changes in community  
16 composition and harmful algal blooms that are indicative of eutrophication. The  
17 following summary is a concise overview of the known or anticipated effects of nitrogen  
18 enrichment on estuaries within the United States.

#### 20 **What is the nature of estuary responses to reactive nitrogen deposition?**

21 *In the ISA, the evidence is sufficient to infer a **causal** relationship between  $N_r$*   
22 *deposition and the biogeochemical cycling of N and carbon (C) in estuaries (**ISA 4.3.4.1***  
23 ***and 3.3.2.3**). In general, estuaries tend to be nitrogen-limited, and many currently receive*  
24 *high levels of nitrogen input from human activities (**REA 5.1.1**). It is unknown if*  
25 *atmospheric deposition alone is sufficient to cause eutrophication; however, the*  
26 *contribution of atmospheric nitrogen deposition to total nitrogen load is calculated for*  
27 *some estuaries and can be >40% (**REA 5.1.1**).*

28 *The evidence is sufficient to infer a **causal** relationship between N deposition and*  
29 *the alteration of species richness, species composition and biodiversity in estuarine*  
30 *ecosystems (**ISA 4.3.4.2 and 3.3.5.4**). Atmospheric and non-atmospheric sources of N*  
31 *contribute to increased phytoplankton and algal productivity, leading to eutrophication.*

1 Shifts in community composition, reduced hypolimnetic DO, decreases in biodiversity,  
2 and mortality of submerged aquatic vegetation are associated with increased N deposition  
3 in estuarine systems.

4

5 **What types of ecosystems are sensitive to such effects? How are these**  
6 **responses affected by atmospheric, ecological, and landscape factors?**

7 Because the productivity of estuarine and near shore marine ecosystems is  
8 generally limited by the availability of N, they are susceptible to the eutrophication effect  
9 of N deposition (**ISA 4.3.4.1**). A recent national assessment of eutrophic conditions in  
10 estuaries found the most eutrophic estuaries were generally those that had large  
11 watershed-to-estuarine surface area, high human population density, high rainfall and  
12 runoff, low dilution, and low flushing rates (Bricker et al., 2007). In the REA, the  
13 National Oceanic and Atmospheric Administration's (NOAA) National Estuarine  
14 Eutrophication Assessment (NEEA) assessment tool, Assessment of Estuarine Tropic  
15 Status (ASSETS) categorical Eutrophication Index (EI) (Bricker et al., 2007) was used to  
16 evaluate eutrophication due to atmospheric loading of nitrogen. ASSETS EI is an  
17 estimation of the likelihood that an estuary is experiencing eutrophication or will  
18 experience eutrophication based on five ecological indicators: chlorophyll *a*, macroalgae,  
19 dissolved oxygen, nuisance/toxic algal blooms and submerged aquatic vegetation (SAV)  
20 (Bricker et al., 2007).

21 In the REA, two regions were selected for case study analysis using ASSETS EI,  
22 the Chesapeake Bay and Pamlico Sound. Both regions received an ASSETS EI rating of  
23 *Bad* indicating that the estuary had moderate to high pressure due to overall human  
24 influence and a moderate high to high eutrophic condition (**REA 5.2.4.1 and 5.2.4.2**).  
25 These results were then considered with SPATIally Referenced Regression (SPARROW)  
26 modeling to develop a response curve to examine the role of atmospheric nitrogen  
27 deposition in achieving a desired decrease in load. To change the Neuse River Estuary's  
28 EI score from *Bad* to *Poor* not only must 100% of the total atmospheric nitrogen  
29 deposition be eliminated, but considerably more nitrogen from other sources as well must  
30 be controlled (REA section 5.2.7.2). In the Potomac River estuary, a 78% decrease of  
31 total nitrogen could move the EI score from *Bad* to *Poor* (**REA 5.2.7.1**). The results of  
32 this analysis indicated decreases in atmospheric deposition alone could not eliminate  
33 coastal eutrophication problems due to multiple non-atmospheric nitrogen inputs (**REA**

1 **7.3.3).** However, by decreasing atmospheric contributions, it may help avoid the need for  
2 more costly controls on nitrogen from other sources. In addition, the somewhat arbitrary  
3 discreteness of the EI scale can mask the benefits of decreases in nitrogen between  
4 categories.

5 In general, estuaries tend to be N-limited (Elser et al., 2008), and many currently  
6 receive high levels of N input from human activities to cause eutrophication (Howarth et  
7 al., 1996; Vitousek and Howarth, 1991). Atmospheric N loads to estuaries in the U.S. are  
8 estimated to range from 2-8% for Guadalupe Bay, TX on the lowest end to as high as  
9 72% for St Catherines-Sapelo estuary, GA (Castro et al., 2003). The Chesapeake Bay is  
10 an example of a large, well-studied and severely eutrophic estuary that is calculated to  
11 receive as much as 30% of its total N load from the atmosphere.

### 12 13 **What is the magnitude of ecosystem responses to eutrophication?**

14 There is a scientific consensus that nitrogen-driven eutrophication in shallow  
15 estuaries has increased over the past several decades and that the environmental  
16 degradation of coastal ecosystems due to nitrogen, phosphorus, and other inputs is now a  
17 widespread occurrence (Paerl et al., 2001). For example, the frequency of phytoplankton  
18 blooms and the extent and severity of hypoxia have increased in the Chesapeake Bay  
19 (Officer et al., 1984) and Pamlico estuaries in North Carolina (Paerl et al., 1998) and  
20 along the continental shelf adjacent to the Mississippi and Atchafalaya rivers' discharges  
21 to the Gulf of Mexico (Eadie et al., 1994).

22 A recent national assessment of eutrophic conditions in estuaries found that 65%  
23 of the assessed systems had moderate to high overall eutrophic conditions and generally  
24 received the greatest N loads from all sources, including atmospheric and land-based  
25 sources (Bricker et al., 2007). Most eutrophic estuaries occurred in the mid-Atlantic  
26 region and the estuaries with the lowest degree of eutrophication were in the North  
27 Atlantic (Bricker et al., 2007). Other regions had mixtures of low, moderate, and high  
28 degrees of eutrophication (**ISA 4.3.4.3**).

29 The mid-Atlantic region is the most heavily impacted area in terms of moderate or  
30 high loss of submerged aquatic vegetation due to eutrophication (**ISA 4.3.4.2**).

31 Submerged aquatic vegetation is important to the quality of estuarine ecosystem habitats  
32 because it provides habitat for a variety of aquatic organisms, absorbs excess nutrients,

1 and traps sediments (**ISA 4.3.4.2**). It is partly because many estuaries and near-coastal  
2 marine waters are degraded by nutrient enrichment that they are highly sensitive to  
3 potential negative impacts from nitrogen addition from atmospheric deposition.  
4

#### 5 **2.2.4 What are the key uncertainties associated with nutrient enrichment?**

6 There are different levels of uncertainty associated with relationships between  
7 deposition, ecological effects and ecological indicators. The criteria used in the REA to  
8 evaluate the degree of confidence in the data, modeling and ecological effect function are  
9 detailed in Chapter 7 of the REA and summarized in section 2.1.4 of this chapter (**REA**  
10 **7.0**).  
11

##### 12 **2.2.4.1 Aquatic**

13 The approach for assessing atmospheric contributions to total nitrogen loading in  
14 the REA, was to consider the main-stem river to an estuary (including the estuary) rather  
15 than an entire estuary system or bay. The biological indicators used in the NOAA  
16 ASSETS EI required the evaluation of many national databases including the US  
17 Geological Survey National Water Quality Assessment (NAWQA) files, EPA's  
18 STORage and RETrival (STORET) database, NOAA's Estuarine Drainage Areas data,  
19 and EPA's water quality standards nutrient criteria for rivers and lakes (**REA Appendix**  
20 **6, Table 1.2.-1**). Both the SPARROW modeling for nitrogen loads and assessment of  
21 estuary conditions under NOAA ASSETS EI, have been applied on a national scale. The  
22 REA concludes that the available data are *medium quality with intermediate confidence*  
23 about the use of these data and their values for extrapolating to a larger regional area  
24 (**REA 7.3.1**). *Intermediate confidence* is associated with the modeling approach using  
25 ASSETS EI and SPARROW. The REA states there is low confidence with the  
26 ecological effect function due to the results of the analysis which indicated that  
27 reductions in atmospheric deposition alone could not solve coastal eutrophication  
28 problems due to multiple non-atmospheric nitrogen inputs (**REA 7.3.3**).  
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1 **2.2.4.2 Terrestrial**

2 Ecological thresholds are identified for CSS and MCF and these data are  
3 considered to be of *high quality*, however, the ability to extrapolate these data to larger  
4 regional areas is limited (**REA 7.4.1**). No quantitative modeling was conducted or  
5 ecological effect function developed for terrestrial nutrient enrichment reflecting the  
6 uncertainties associated with these depositional effects.

7  
8 **2.3 WHAT ECOLOGICAL EFFECTS ARE ASSOCIATED WITH GAS-  
9 PHASE NO<sub>x</sub> AND SO<sub>x</sub>?**

10 Acidifying deposition and nitrogen enrichment are the main focus of this policy  
11 assessment; however, there are other known ecological effects are attributed to gas-phase  
12 NO<sub>x</sub> and SO<sub>x</sub>. Acute and chronic exposures to gaseous pollutants such as sulfur dioxide  
13 (SO<sub>2</sub>), nitrogen dioxide (NO<sub>2</sub>), nitric oxide (NO), nitric acid (HNO<sub>3</sub>) and peroxyacetyl  
14 nitrite (PAN) are associated with negative impacts to vegetation. The current secondary  
15 NAAQS were set to protect against direct damage to vegetation by exposure to gas-phase  
16 NO<sub>x</sub> or SO<sub>x</sub>, such as foliar injury, decreased photosynthesis, and decreased growth. The  
17 following summary is a concise overview of the known or anticipated effects to  
18 vegetation caused by gas phase N and S. Most phototoxic effects associated with gas  
19 phase NO<sub>x</sub> and SO<sub>x</sub> occur at levels well above ambient concentrations observed in the  
20 U.S. (**ISA 3.4.2.4**).

21  
22 **2.3.1 What is the nature of ecosystem responses to gas-phase nitrogen and sulfur?**

23 The 2008 ISA found that gas phase N and S are associated with direct phytotoxic  
24 effects (**ISA 4.4**). *The evidence is sufficient to infer a **causal** relationship between*  
25 *exposure to SO<sub>2</sub> and injury to vegetation (**ISA 4.4.1 and 3.4.2.1**)*. Acute foliar injury to  
26 vegetation from SO<sub>2</sub> may occur at levels above the current secondary standard (3-h  
27 average of 0.50 ppm). Effects on growth, reduced photosynthesis and decreased yield of  
28 vegetation are also associated with increased SO<sub>2</sub> exposure concentration and time of  
29 exposure.

30 *The evidence is sufficient to infer a **causal** relationship between exposure to NO,*  
31 *NO<sub>2</sub> and PAN and injury to vegetation (**ISA 4.4.2 and 3.4.2.2**)*. At sufficient

1 concentrations, NO, NO<sub>2</sub> and PAN can decrease photosynthesis and induce visible foliar  
2 injury to plants. *Evidence is also sufficient to infer a **causal** relationship between*  
3 *exposure to HNO<sub>3</sub> and changes to vegetation (ISA 4.4.3 and 3.4.2.3).* Phytotoxic effects  
4 of this pollutant include damage to the leaf cuticle in vascular plants and disappearance of  
5 some sensitive lichen species.

6 **2.3.2 What types of ecosystems are sensitive to such effects? How are these**  
7 **responses affected by atmospheric, ecological, and landscape factors?**

8 Vegetation in ecosystems near sources of gaseous NO<sub>x</sub> and SO<sub>x</sub> or where  
9 ambient concentrations of SO<sub>2</sub>, NO, NO<sub>2</sub>, PAN and HNO<sub>3</sub> are higher are more likely to  
10 be impacted by these pollutants. Uptake of these pollutants in a plant canopy is a complex  
11 process involving adsorption to surfaces (leaves, stems and soil) and absorption into  
12 leaves (ISA 3.4.2). The functional relationship between ambient concentrations of gas  
13 phase NO<sub>x</sub> and SO<sub>x</sub> and specific plant response are impacted by internal factors such as  
14 rate of stomatal conductance and plant detoxification mechanisms, and external factors  
15 including plant water status, light, temperature, humidity, and pollutant exposure regime  
16 (ISA 3.4.2).

17 Entry of gases into a leaf is dependent upon physical and chemical processes of  
18 gas phase as well as to stomatal aperture. The aperture of the stomata is controlled  
19 largely by the prevailing environmental conditions, such as water availability, humidity,  
20 temperature, and light intensity. When the stomata are closed, resistance to gas uptake is  
21 high and the plant has a very low degree of susceptibility to injury. Mosses and lichens  
22 do not have a protective cuticle barrier to gaseous pollutants or stomata and are generally  
23 more sensitive to gaseous sulfur and nitrogen than vascular plants (ISA 3.4.2).

24 The appearance of foliar injury can vary significantly across species and growth  
25 conditions affecting stomatal conductance in vascular plants (REA 6.4.1). For example,  
26 damage to lichens from SO<sub>2</sub> exposure include decreases in photosynthesis and  
27 respiration, damage to the algal component of the lichen, leakage of electrolytes,  
28 inhibition of nitrogen fixation, decreased K<sup>+</sup> absorption, and structural changes (Belnap  
29 et al., 1993; Farmer et al., 1992, Hutchinson et al., 1996).

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**2.3.3 What is the magnitude of ecosystem responses to gas phase effects of NOx and SOx?**

The phytotoxic effects of gas phase NOx and SOx are dependent on the exposure concentration and duration and species sensitivity to these pollutants. Effects to vegetation associated with NOx and SOx, are therefore, variable across the U.S. and tend to be higher near sources of photochemical smog. For example, SO<sub>2</sub> is considered to be the primary factor contributing to the death of lichens in many urban and industrial areas, with fruticose lichens being more susceptible to SO<sub>2</sub> than many foliose and crustose species (Hutchinson et al., 1996).

The ISA states there is very limited new research on phytotoxic effects of NO, NO<sub>2</sub>, PAN and HNO<sub>3</sub> at concentrations currently observed in the United States with the exception of some lichen species (**ISA 4.4**). Past and current HNO<sub>3</sub> concentrations may be contributing to the decline in lichen species in the Los Angeles basin (Boonpragob and Nash 1991; Nash and Sigal, 1999; Riddell et al., 2008). PAN is a very small component of nitrogen deposition in most areas of the United States (**REA 6.4.2**). Current deposition of HNO<sub>3</sub> is contributing to N saturation of some ecosystems close to sources of photochemical smog (Fenn et al., 1998) such as the MCF's of the Los Angeles basin mountain (Bytnerowicz et al., 1999). Most phototoxic effects associated with gas phase NOx and SOx occur at levels well above ambient concentrations observed in the U.S. (**ISA 3.4.2.4**).

**2.4 SUMMARY**

In summary, NOx and SOx in the atmosphere contribute to effects on individual species and ecosystems through direct contact with vegetation, and more significantly through deposition to sensitive ecosystems. The ISA concludes that the evidence is sufficient to conclude causal relationships between acidifying deposition of N and S and effects on freshwater aquatic ecosystems and terrestrial ecosystems, and between nitrogen nutrient enrichment and effects on sensitive terrestrial and freshwater aquatic ecosystems. The ISA also concludes that a causal relationship is supported between nitrogen nutrient enrichment and effects on estuarine ecosystems; however, the

1 contribution of atmospheric oxidized nitrogen relative to reduced nitrogen and non-  
2 atmospheric nitrogen is more difficult to determine.

3         The REA provides additional support that under recent conditions, deposition  
4 levels have exceeded benchmarks for ecological indicators of acidification and nutrient  
5 enrichment that indicate that effects are likely to be widespread in lakes and streams  
6 within sensitive ecosystems.

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1           **3   CONSIDERATIONS OF ADVERSITY TO PUBLIC WELFARE**

2  
3   **3.1   How do we characterize adversity to public welfare? What are the relevant factors**  
4   **and how are they addressed in this document?**

5  
6           Characterizing a known or anticipated adverse effect to public welfare is an important  
7 component of developing any secondary NAAQS. According to the Clean Air Act, welfare  
8 effects include:

9           effects on soils, water, crops, vegetation, manmade materials, animals, wildlife,  
10          weather, visibility, and climate, damage to and deterioration of property, and  
11          hazards to transportation, as well as effect on economic values and on personal  
12          comfort and well-being, whether caused by transformation, conversion, or  
13          combination with other air pollutants (CAA, Section 302(h)).  
14

15          While the text above lists a number of welfare effects, these effects do not define public  
16 welfare in and of themselves.

17           Although there is no specific definition of adversity to public welfare, the paradigm of  
18 adversity to public welfare as deriving from disruptions in ecosystem structure and function has  
19 been used broadly by EPA to categorize effects of pollutants from the cellular to the ecosystem  
20 level. An evaluation of adversity to public welfare might consider the likelihood, type,  
21 magnitude, and spatial scale of the effect as well as the potential for recovery and any  
22 uncertainties relating to these considerations.

23           Similar concepts were used in past reviews of secondary NAAQS for ozone, PM (relating  
24 to visibility), as well as in initial reviews of effects from lead deposition. Because NO<sub>y</sub> and SO<sub>x</sub>  
25 are deposited from ambient sources into ecosystems where they affect changes to organisms,  
26 populations and ecosystems, the concept of adversity to public welfare as related to impacts on  
27 the public from alterations in structure and function of ecosystems is appropriate for this review.  
28 Other information that may be helpful to consider includes the role of critical loads and  
29 ecosystem service impacts as benchmarks or measures of impacts on ecosystems that may affect  
30 public welfare. Ecosystem services can be related directly to concepts of public welfare to  
31 inform discussions of societal adverse impacts. In subsequent sections we will discuss each of  
32 these concepts as they relate to adversity.  
33  
34

1 **3.1.1 What are the benchmarks for adversity from other sources?**

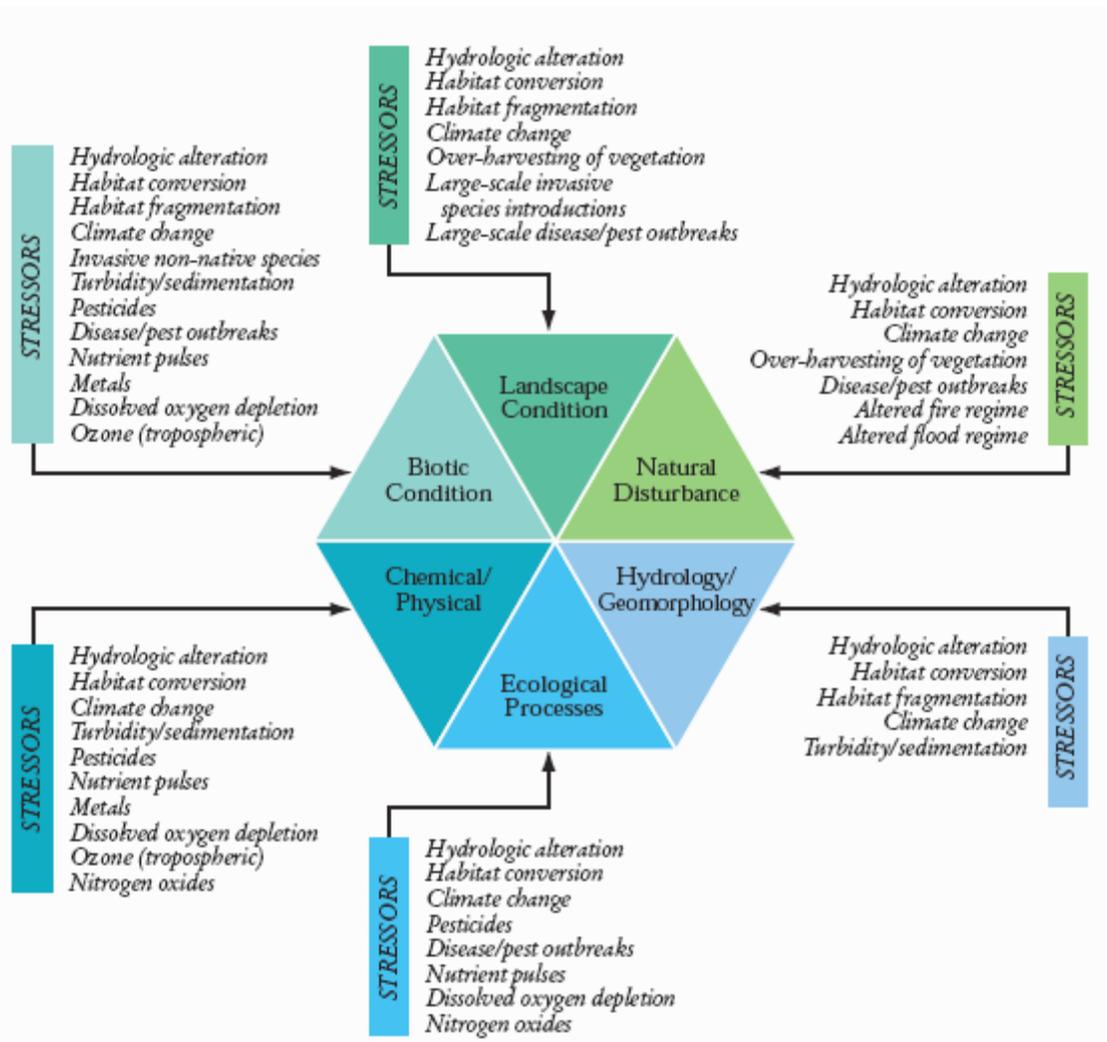
2 **3.1.1.1 Ozone NAAQS Review**

3 The evaluation of adversity from a public welfare perspective in the context of ozone and  
4 particulate matter (PM) are relevant to this current review. Both ozone and PM have  
5 documented effects on ecological receptors. These criteria pollutants are being reviewed on a  
6 schedule as part of the NAAQS process. The ozone secondary standard is currently under  
7 reconsideration from the 2008 ruling with a proposal was published January 6, 2010. The final  
8 Policy Assessment for PM is being developed and is expected to be finalized in the fall of 2010.

9  
10 For the purposes of the reconsideration of the secondary standard for ozone welfare  
11 effects of ozone are primarily limited to vegetation. These effects begin at the level of the  
12 individual cell and accumulate up to the level of whole leaves and plants. If effects occur on  
13 enough individual plants within the population, communities and ecosystems may be impacted.  
14 Prior to the 2008 ozone review, Ozone vegetation effects were classified as either “injury” or  
15 “damage” (FR 72 37889). “Injury” was defined as; encompassing all plant reactions, including  
16 reversible changes or changes in plant metabolism, quality or reduced growth that does not  
17 impair the intended use of the plant while “damage” includes those injury effects that reach  
18 sufficient magnitude as to decrease or impair the intended use of the plant (FR 72 37890). The  
19 “intended use” of the plant was imbedded with the concept of adversity to public welfare.  
20 Ozone-associated “damage” was considered adverse if the intended use of the plant was  
21 compromised (i.e. crops, ornamentals, plants located in Class I areas). Effects of ozone on  
22 single plants or species grown in monocultures such as agricultural crops and managed forests  
23 were evaluated without consideration of potential effects on natural forests or entire ecosystems.

24 In the 2008 rulemaking, EPA expanded the characterization of adversity beyond the  
25 individual plant level and this language is continued in the 2010 ozone reconsideration. The  
26 2008 final rule and 2010 proposal conclude that a determination of what constitutes an “adverse”  
27 welfare effect in the context of secondary NAAQS review can appropriately occur by  
28 considering effects at higher ecological levels (populations, communities, ecosystems) as  
29 supported by recent literature. The ozone review uses the example of the construct presented in  
30 Hogsett et al. (1997) as a model for assessing risks to forests. This study suggests that adverse  
31 effects could be classified into one or more of the following categories: (1) economic production,

1 (2) ecological structure, (3) genetic resources, and (4) cultural values”. Another recent  
 2 publication, “A Framework for Assessing and Reporting on Ecological Condition: an SAB  
 3 report” (Young and Sanzone, 2002) provides additional support for expanding the consideration  
 4 of adversity beyond the species level and at higher levels by making explicit the linkages  
 5 between stress-related effects at the species level and at higher levels within an ecosystem  
 6 hierarchy (See Figure 3-1).



7  
 8 **Figure 3-1. Common anthropogenic stressors and the essential ecological attributes they**  
 9 **affect.** Modified from Young and Sanzone (2002)  
 10

11 In the 2008 ozone NAAQS review and current ozone NAAQS proposal, the  
 12 interpretation of what constitutes an adverse effect on public welfare can vary depending on the  
 13 location and intended use of the plant. The degree to which O<sub>3</sub>-related effects are considered  
 14 adverse to public welfare depends on the intended use of the vegetation and its significance to

1 public welfare (73 FR 16496). Therefore, effects on vegetation (e.g., biomass loss, foliar injury,  
2 impairment of intended use) may be judged to have a different degree of impact on public  
3 welfare depending, for example, on whether that effect occurs in a Class I area, a city park,  
4 commercial cropland or private land.

5 In the proposed ozone reconsideration in 2010 the Administrator has found that the types  
6 of information most useful in informing the selection of an appropriate range of protective levels  
7 is appropriately focused on information regarding exposures and responses of sensitive trees and  
8 other native species that occur in protected areas such as Class I areas or on lands set aside by  
9 States, Tribes and public interest groups to provide similar benefits to the public welfare. She  
10 further notes that while direct links between O<sub>3</sub> induced visible foliar injury symptoms and other  
11 adverse effects (e.g., biomass loss) are not always found, visible foliar injury in itself is  
12 considered by the National Park Service (NPS) to affect adversely air quality related values  
13 (AQRV) in Class I areas. while the Administrator recognizes that uncertainty remains as to what  
14 level of annual tree seedling biomass loss when compounded over multiple years should be  
15 judged adverse to the public welfare, she believes that the potential for such anticipated effects  
16 should be considered in judging to what degree a standard should be precautionary (73 FR  
17 16496). The range of proposed levels from 7 – 15 ppb includes at the maximum level of 15 ppb  
18 protection of approximately 75% of seedlings from more than 10% biomass loss.

### 19 20 **3.1.2 Other EPA Programs and Federal Agencies**

21 Various federal laws and policies exist to protect ecosystem health. How other federal  
22 agencies and EPA offices consider ecosystem effects in carrying out their programs can help  
23 inform the Administrator when she evaluates the adversity of ecosystem impacts on public  
24 welfare. From the 1996 National Acid Precipitation Assessment Program Report to Congress: “  
25 The 1990 Clean Air Act Amendments require that the National Acid Precipitation Assessment  
26 Program (NAPAP) prepare biennial reports to Congress, and that “every four years ... the report  
27 ... shall include the reduction in deposition rates that must be achieved in order to prevent  
28 adverse ecological effects” (Public Law 101-549, Title IX, Section 903 (j)(3)(F)(i), codified as  
29 amended at 42 USC §7403(j)(3)(F)(I)). Although the term *adverse ecological effects* is not  
30 specifically defined in the Clean Air Act Amendments, a working definition can be derived from  
31 relevant statements at various locations in the statute. Congress expresses its concern with

1 ecological components (the scope is broad and inclusive, since ecology encompasses the  
2 interrelationships of organisms and their environment) in the preceding subsection (E) of the  
3 statute. That subsection requires reporting on “the status of ecosystems (including forest and  
4 surface waters) ... affected by acid deposition ... including changes in surface water quality and  
5 forest and soil conditions ... [and] high elevation watersheds” (42 USC §7403(j)(3)(E)(iii-v)).  
6 The adverse effects of concern to Congress, as evidenced in its findings and declaration of  
7 purpose, are the “dangers to the public health and welfare ... including injury ... damage ... and  
8 ... deterioration” (42 USC §7401(a)). Based on the intent of Congress, as expressed above and  
9 elsewhere in the Clean Air Act, and shaped by indications of intent expressed in other relevant  
10 environmental statutes and regulations, the following working definition of *adverse ecological*  
11 *effects* has been derived and is used in the preparation of the NAPAP report:

12 any injury (i.e., loss of chemical or physical quality or viability) to any  
13 ecological or ecosystem component, up to and including at the regional  
14 level, over both long and short terms. Similarly, adverse effects for other  
15 areas of concern addressed in this report—i.e., visibility, materials, and  
16 human health—consist of loss of quality up to and including at the  
17 regional level, over both long and short terms.”  
18

19 As another example, an effect may be considered adverse to *public welfare* if it  
20 contributes to the inability of areas to meet water quality objectives as defined by the Clean  
21 Water Act. The following federal statutes and policies may prove helpful to consider.

22

### 23 **3.1.21. Prevention of Significant Deterioration Program**

24 The Clean Air Act’s Prevention of Significant Deterioration (PSD) program (42 U.S.C.  
25 7470) purposes include to “preserve, protect and enhance the air quality in national parks,  
26 wilderness areas and other areas of natural, recreational, scenic or historic value . . . .” Also, the  
27 PSD program charges the Federal Land Managers, including the NPS, with “. . . an affirmative  
28 responsibility to protect the air quality related values . . . “within federal Class I lands. (42 U.S.C.  
29 7475(d)(2)(B)).

30

31

1  
2           **3.1.2.2 EPA Office of Water**

3           Section 101 of the Clean Water Act (CWA) (Declaration of Goals and Policy) states that  
4 the objective of the CWA is to restore and maintain the chemical, physical, and biological  
5 integrity of the Nation’s waters and to attain, where possible, water quality that protects fish,  
6 shellfish, wildlife and provides for water-based recreation.

7           The CWA also authorizes EPA to develop water quality criteria as a guide for the states  
8 to set water quality standards to protect aquatic life. In consideration of acidification effects,  
9 EPA’s Redbook, *Quality Criteria for Water*, published originally in 1976, recommends that  
10 alkalinity be 20 mg/l/L or more as CaCO<sub>3</sub> for freshwater aquatic life except where natural  
11 concentrations are less. Alkalinity is the sum total of components in the water that tend to  
12 elevate the pH of the water above a value of about 4.5.

13           As mentioned in the Redbook, alkalinity is expressed as CaCO<sub>3</sub> in mg/L. Alkalinity  
14 differs slightly from ANC in that ANC includes other buffering compounds (Na, Mg, and K) as  
15 well and includes buffering capacity of particulates in the water sample. Since alkalinity is  
16 expressed as mg/L and ANC is expressed as µeq/L, alkalinity must be multiplied by 20 to be  
17 converted to µeq/L. Thus a recommended criterion of 20 mg/L alkalinity is equivalent to an  
18 ANC of 400 µeq/L.

19  
20           **3.1.2.3 National Park Service**

21           The National Park Service (NPS) is responsible for the protection of all resources within  
22 the national park system. These resources include those that are related to and/or dependent  
23 upon good air quality, such as whole ecosystems and ecosystem components. The NPS, in its  
24 Organic Act (16 U.S.C. 1), is directed to conserve the scenery, natural and historic objects and  
25 wildlife and to provide for the enjoyment of these resources unimpaired for current and future  
26 generations.

27           The Wilderness Act of 1964 asserts wilderness areas will be administered in such a  
28 manner as to leave them unimpaired and preserve them for the enjoyment of future generations.

29           NPS Management Policies (2006) guide all NPS actions including natural resources  
30 management. In general, the NPS Management Policies reiterate the NPS Organic Act’s  
31 mandate to manage the resources “unimpaired.”

1           **3.1.2.3           U.S. Fish and Wildlife Service**

2           On endangered species, Title 16 USC Chapter 35 Section 1531 states “The Congress  
3 finds and declares that— these species of fish, wildlife , and plants are of esthetic , ecological,  
4 educational, historical, recreational, and scientific value to the Nation and its people and that all  
5 Federal departments and agencies will use their authorities to conserve threatened and  
6 endangered species.

7           The United States Fish and Wildlife Service (FWS) manages the National Wildlife  
8 Refuge System lands to “...ensure that the biological integrity, diversity, and environmental  
9 health of the Systems are maintained for the benefit of present and future generations of  
10 Americans.” 16 U.S.C. Section 668dd(a)(4)(B)(1997).

11  
12           **3.1.2.4           U.S. Forest Service**

13           The National Forest units are managed consistent with Land and Resource Management  
14 Plans (LRMPs) under the provisions of the National Forest Management Act (NFMA). 16  
15 §U.S.C. 1604 (1997). LRMPs are, in part, specifically based on recognition that the National  
16 Forests are ecosystems and their management for goods and services requires an awareness and  
17 consideration of the interrelationships among plants, animals, soil, water, air, and other  
18 environmental factors within such ecosystems. 36 C.F.R. §219.1(b)(3)

19           Any measures addressing Air Quality Related Values (AQRV) on National Forest  
20 System lands will be implemented through, and be consistent with, the provisions of an  
21 applicable LRMP or its revision (16 U.S.C. §1604(i)). Additionally, the Secretary of Agriculture  
22 must prepare a Renewable Resource Program that recognizes the need to protect and, if  
23 necessary, improve the quality of air resources. 16 U.S.C. §1602(5)(C).

24           AQRVs in Wilderness areas may receive further protection by the previously mentioned  
25 1964 Wilderness Act. For Wilderness Areas in the National Forest System, the Act's  
26 implementing regulations are found at 36 C.F.R. §293 requiring these Wilderness Areas be  
27 administered to preserve and protect [their] wilderness character.

28  
29           **3.1.25 Chesapeake Bay Total Maximum Daily Loads**

30           Under section 303(d) of the Clean Water Act, states, territories, and authorized tribes are  
31 required to develop lists of impaired waters. These are waters that are too polluted or otherwise

1 degraded to meet the water quality standards set by states, territories, or authorized tribes. The  
2 law requires that these jurisdictions establish priority rankings for waters on the lists and develop  
3 TMDLs for these waters. A Total Maximum Daily Load, or TMDL, is a calculation of the  
4 maximum amount of a pollutant that a waterbody can receive and still safely meet water quality  
5 standards. EPA is developing a TMDL for the Chesapeake Bay and its tributaries. The  
6 Chesapeake Bay Program has modeled the level of nitrogen that can reach the Bay and still meet  
7 the Bay's water quality standards. The TMDL, with full public participation, will set waste load  
8 allocations for point source discharges and load allocations for nonpoint sources of nitrogen. Air  
9 deposition to the Bay and its watershed, as a source category, will have a specific allocation.  
10 The allocation can be used to calculate the level of ambient air concentrations of reactive  
11 nitrogen that are likely to meet the deposition allocation. To find the NO<sub>y</sub> portion of the  
12 allocation one would subtract the reduced forms from the total allocation. If the total load to the  
13 Bay of nitrogen from all the allocated source categories remains below the allocations, then the  
14 Bay is expected to meet the water quality standards, which are set to protect the designated uses  
15 of the Bay. Since the designated uses are set by the states with public input, not meeting the  
16 designated uses can be seen as having an adverse effect to public welfare.

17

### 18 **3.1.2.6 Critical Loads**

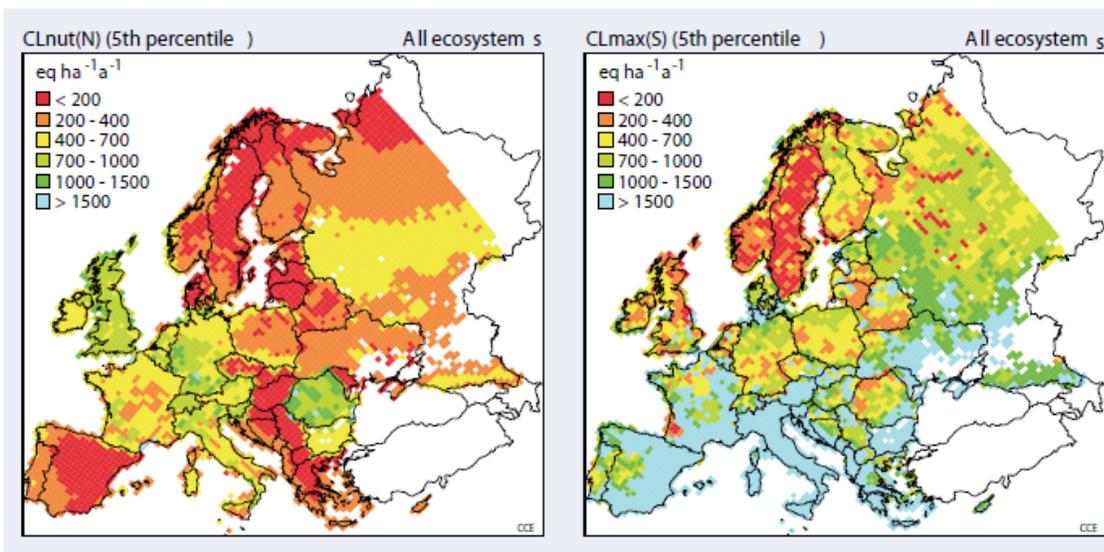
19 The term critical load is used to describe the threshold of air pollution deposition that  
20 causes a specified level of harm to sensitive resources in an ecosystem. A critical load is  
21 technically defined as “the quantitative estimate of an exposure to one or more pollutants below  
22 which significant harmful effects on specified sensitive elements of the environment are not  
23 expected to occur according to present knowledge” (Nilsson and Grennfelt, 1988). The  
24 determination of when a harmful effect becomes “significant” may be in the view of a researcher  
25 or through a policy development process. Researchers often use the term “critical loads” to  
26 describe when particular detrimental effects are realized, as is the case in Figure 2-1.

27 Harmful effects due to acidification have been defined here as those that occur below a  
28 given ANC for aquatic systems and below a given Bc:Al ratio for terrestrial systems. However,  
29 the level at which an effect becomes adverse to public welfare is determined by the  
30 Administrator, informed by available scientific information.

31

### 3.1.2.7 United Nations Economic Commission for Europe (UNECE)

In many European countries a critical loads framework is used to determine a level of damages to ecosystem services from pollution that is legally allowed. These critical loads are determined through a policy process. Indeed critical loads have been modeled by individual countries and submitted to the UNECE (in cases where countries have not submitted their own critical loads those loads have been calculated for them) and are being used to support international emissions reduction agreements including the 1999 Gothenburg protocol and the National Emission Ceiling Directive of the European Commission. Figure 3-2 shows critical loads for eutrophication and acidification that protect 95% of forests, seminatural vegetation or surface waters in Europe. For comparison to the U.S maps presented in this document the units of deposition convert to a range of 3.2 kg ha<sup>-1</sup>a<sup>-1</sup> to > 24 kg ha<sup>-1</sup>a<sup>-1</sup>.



**Figure 3-2 European maps of eutrophication (left) and acidification (right) which protect 95% of natural areas in 50x50 km<sup>2</sup> European Monitoring and Evaluation Programme grid. Red shaded areas illustrate grid cells where deposition needs to be lower than 200 eq ha<sup>-1</sup> a<sup>-1</sup> to reach this protection target.**

Source: Critical Load, Dynamic Modelling and Impact Assessment in Europe CCE Status Report 2008 available at <http://www.pbl.nl/en/publications/2009/Critical-load-dynamic-modelling-and-impact-assessment-in-Europe-CCE-Status-Report-2008.html>

The Coordination Center for Effects, a working center for the Working Group on Effects of the Convention on Long Range Transboundary Pollution, in the 2008 status report shows calculated critical loads based on an ANC target of 20 µeq/L for the protection of brown trout.

1 Individual countries have also set ANC targets for other species for example Norway uses a  
2 critical limit of 30µeq/L for Atlantic salmon (Jenkins et al, 2003).

3

### 4 **3.2 What are ecosystem services and how does this concept relate to public welfare?**

5 An additional concept that may be useful in considering the issue of adversity to public  
6 welfare is ecosystem services. In the next section the concept of ecosystem services, its  
7 relationship to adversity and public welfare within the context of this review are explained.

8 Ecosystem services can be generally defined as the benefits individuals and organizations  
9 obtain from ecosystems. Ecosystem services can be classified as provisioning (food and water),  
10 regulating (control of climate and disease), cultural (recreational), and supporting (nutrient  
11 cycling) (MEA 2005). Conceptually, changes in ecosystem services may be used to aid in  
12 characterizing a known or anticipated adverse effect to public welfare. In the context of this  
13 review, ecosystem services may also aid in assessing the magnitude and significance to the  
14 public of a resource and in assessing how NO<sub>y</sub> and SO<sub>x</sub> concentrations and deposition may  
15 impact that resource. The relationship between ecosystem services and public welfare effects is  
16 illustrated in Table 3-1.

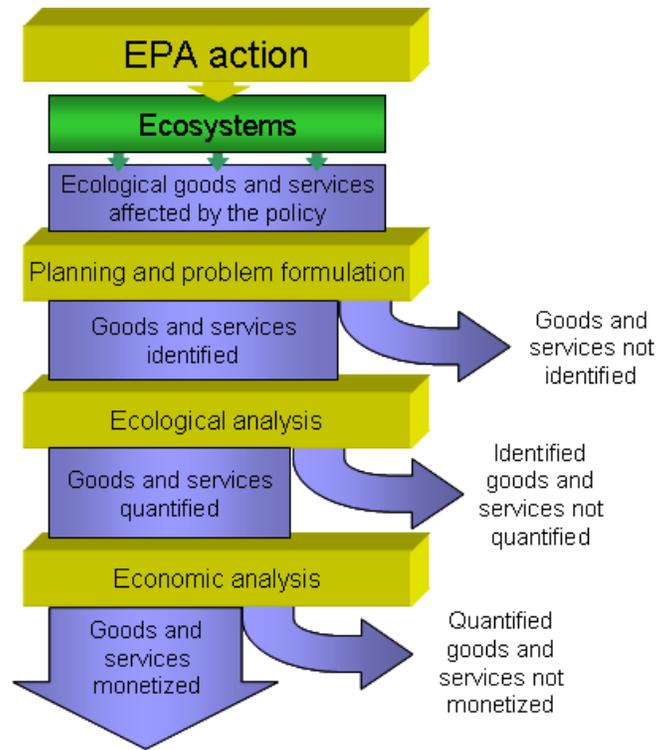
1 **Table 3-1. Crosswalk between Ecosystem Services and Public Welfare Effects**

Public Welfare Effect	Ecosystem Service	Service Category
Soils	Nutrient Cycling	Supporting
Water	Drinking water, Recreation, Aesthetic	Provisioning, Cultural
Crops	Food, Fuel Production, Forest Products	Provisioning
Vegetation	Food, Recreation, Aesthetic, Nonuse	Provisioning, Cultural
Wildlife	Recreation, Food, Nonuse	Cultural, Provisioning
Climate	Climate Control	Regulating
*Personal Comfort and Wellbeing		

2 \*All ecosystem services contribute to personal comfort and wellbeing.

3  
 4 EPA has defined ecological goods and services for the purposes of a Regulatory Impact  
 5 Analysis as the “outputs of ecological functions or processes that directly or indirectly contribute  
 6 to social welfare or have the potential to do so in the future. Some outputs may be bought and  
 7 sold, but most are not marketed” (US EPA 2006). Additionally Executive Order 12866 requires  
 8 a regulatory Impact Analysis for any rule considered “economically significant” and defines  
 9 significant as a rule having \$100 million or more in impacts. Though this is not a definition  
 10 specifically for use in the NAAQS process it may be a useful one in considering the scope of  
 11 ecosystem services and the effects of air pollutants upon those services. Especially important is  
 12 the acknowledgement that it is difficult to measure and/or monetize the goods and services  
 13 supplied by ecosystems. Valuing ecological benefits, or the contributions to social welfare  
 14 derived from ecosystems, can be challenging as noted in EPA’s Ecological Benefits Assessment  
 15 Strategic Plan (US EPA 2006) and the Science Advisory Board report “Valuing the Protection of  
 16 Ecological Systems and Services” (US EPA, 2009). It can be informative in characterizing  
 17 adversity to public welfare to attempt to place an economic valuation on the set of goods and

1 services that have been identified with respect to a change in policy however it must be noted  
2 that this valuation will be incomplete and illustrative only. The stepwise concept leading to the  
3 valuation of ecosystem services is graphically depicted in Figure 3-3.



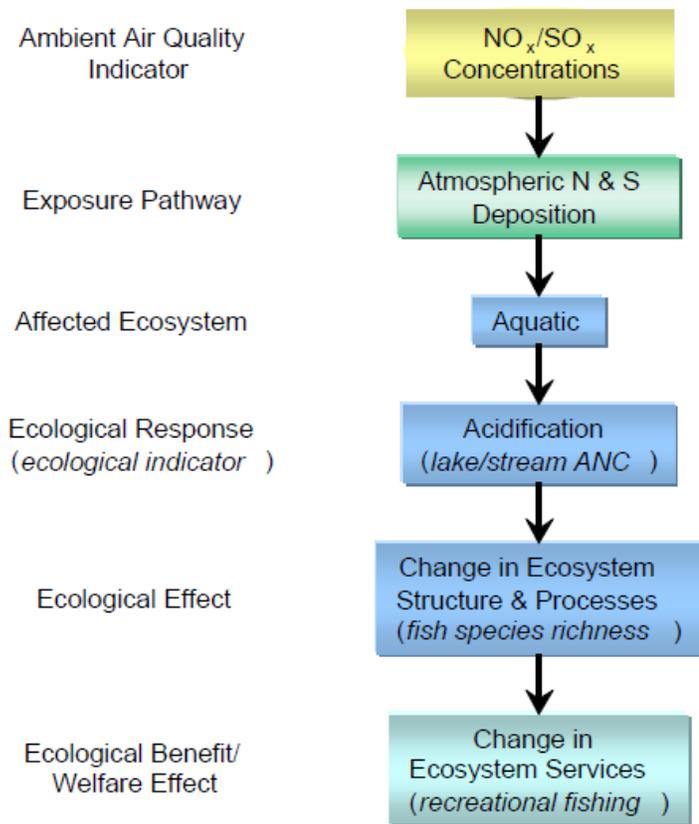
20 **Figure 3-3. Representation of the benefits assessment process indicating where some**  
21 **ecological benefits may remain unrecognized, unquantified, or unmonetized.**  
22 **(Source: EBASP USEPA 2006).**  
23

24 A conceptual model integrating the role of ecosystem services in characterizing known or  
25 anticipated adverse effects to public welfare is shown in Figure 3-3. Under Section 109 of the  
26 CAA, the secondary standard is to specify a level of air quality that is requisite to protect public  
27 welfare. For this review, the relevant air quality indicator is interpreted as ambient  $\text{NO}_y$  and  $\text{SO}_x$   
28 concentrations that can be linked to levels of deposition for which there are ecological effects  
29 that are adverse to public welfare. The case study analyses (described in Chapters 4 and 5 of the  
30 REA and summarized in Chapter 2 of this document) link deposition in sensitive ecosystems  
31 (e.g., the exposure pathway) to changes in a given ecological indicator (e.g., for aquatic  
32 acidification, changes in acid neutralizing capacity [ANC]) and then to changes in ecosystems

1 and the services they provide (e.g., fish species richness and its influence on recreational  
2 fishing). To the extent possible for each targeted effect area, ambient concentrations of nitrogen  
3 and sulfur (i.e., ambient air quality indicators) were linked to deposition in sensitive ecosystems  
4 (i.e., exposure pathways), and then deposition was linked to system response as measured by a  
5 given ecological indicator (e.g., lake and stream acidification as measured by ANC). The  
6 ecological effect (e.g., changes in fish species richness) was then, where possible, associated  
7 with changes in ecosystem services and their public welfare effects (e.g., recreational fishing).  
8 We recognize that there is a certain amount of natural change in ecosystems over time that can  
9 affect the level of acidity and the response of the ecosystem to additional acid and nutrient  
10 inputs. However, this review is focused on the impact of anthropogenic nitrogen and sulfur  
11 deposition given the existing state of non-anthropogenically determined ecosystem  
12 characteristics, and as such we essentially hold these other factors as “fixed” for the purposes of  
13 the review.

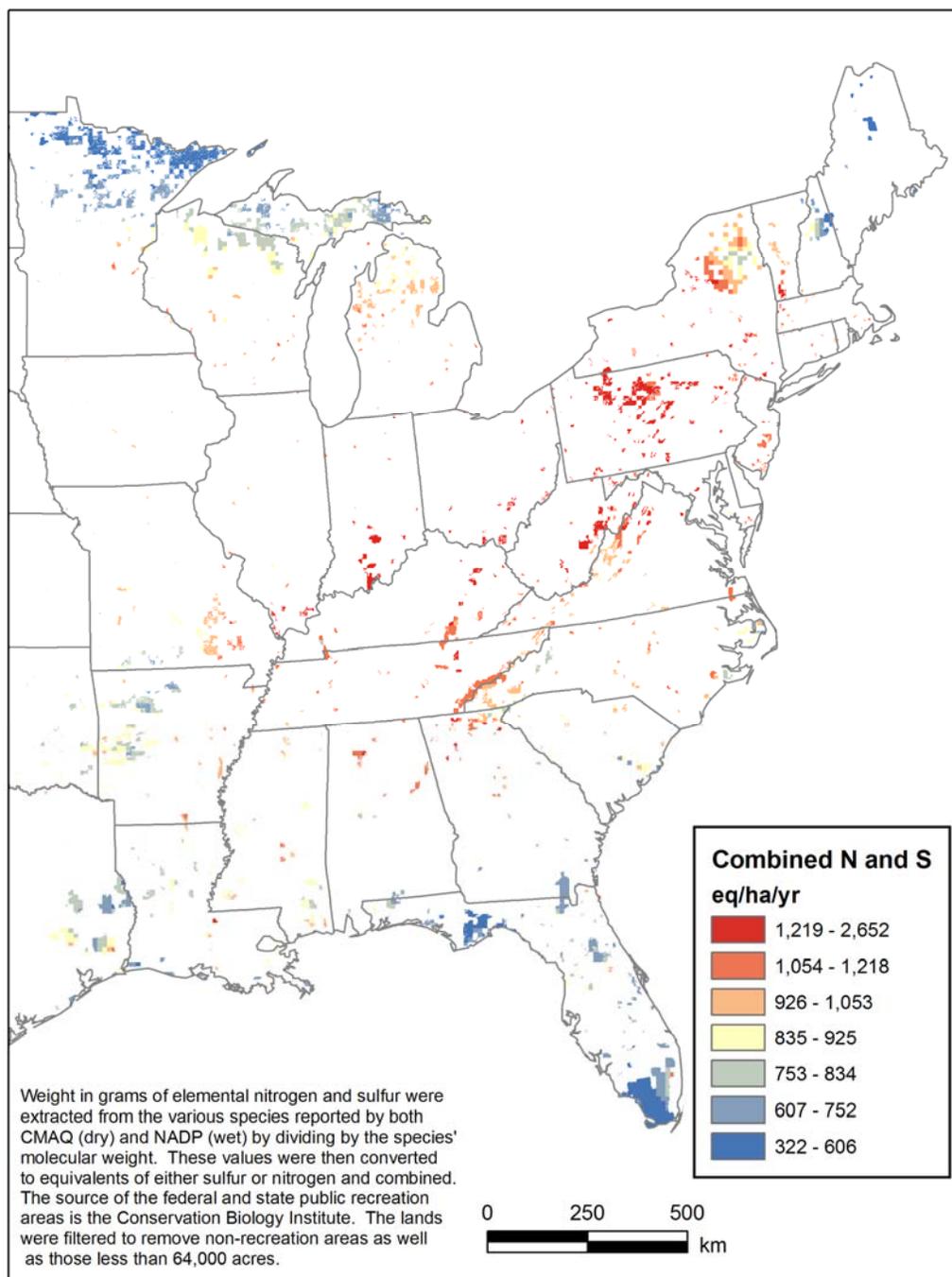
14 Knowledge about the relationships linking ambient concentrations and ecosystem  
15 services can be used to inform a policy judgment on a known or anticipated adverse public  
16 welfare effect. The conceptual model outlined for aquatic acidification in Figure 3-3 can be  
17 modified for any targeted effect area where sufficient data and models are available. For  
18 example, a change in an ecosystem structure and process, such as foliar injury, would be  
19 classified as an ecological effect, with the associated changes in ecosystem services, such as  
20 primary productivity, food availability, forest products, and aesthetics (e.g., scenic viewing),  
21 classified as public welfare effects. Additionally, changes in biodiversity would be classified as  
22 an ecological effect, and the associated changes in ecosystem services—productivity,  
23 recreational viewing and aesthetics—would be classified as public welfare effects. This  
24 information can then be used by the Administrator to determine whether or not the changes  
25 described are adverse to public welfare. In subsequent sections these concepts are applied to  
26 characterize the ecosystem services potentially affected by nitrogen and/or sulfur for each of the  
27 effect areas assessed in the REA.

28

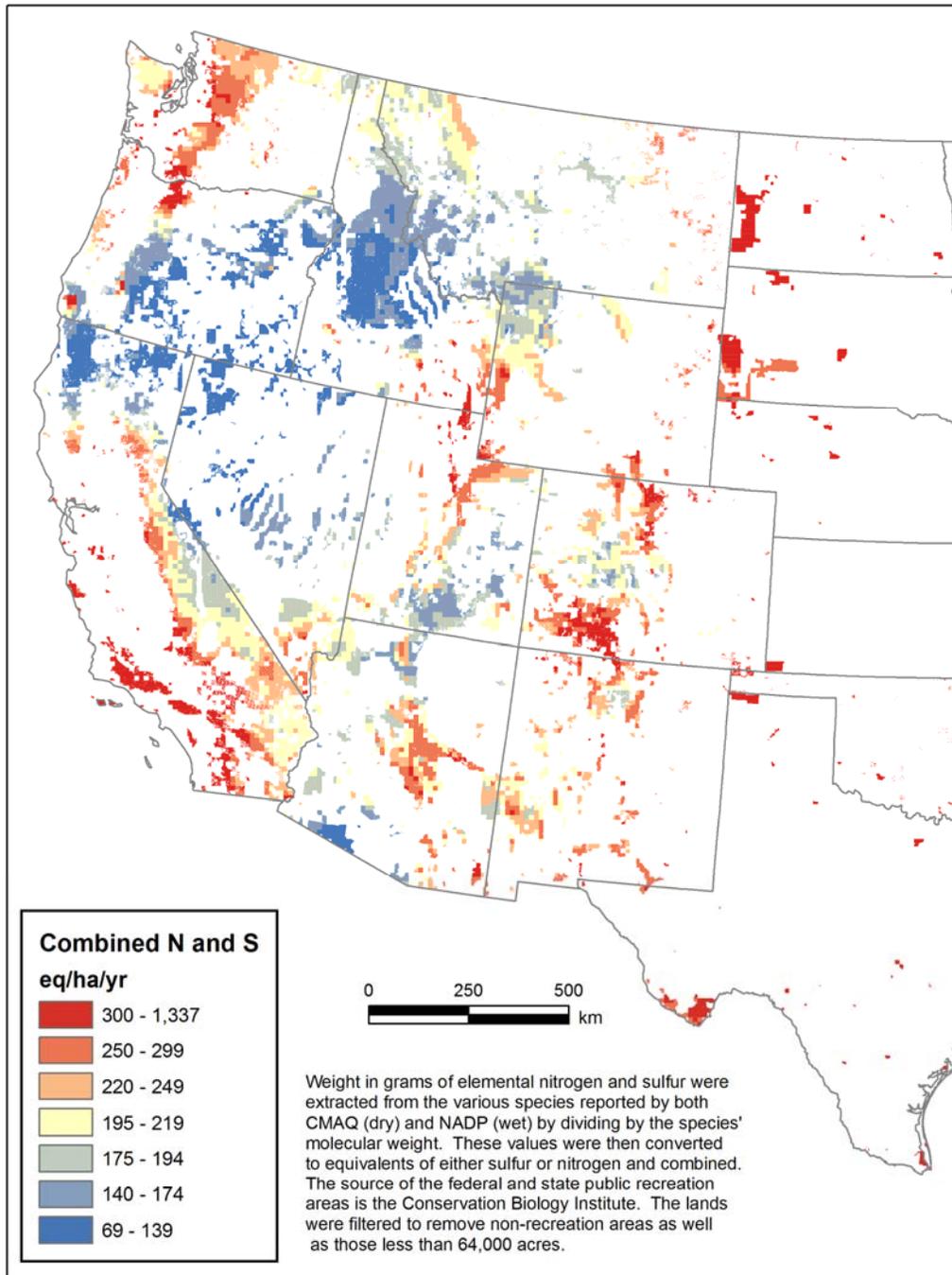


1  
 2 **Figure 3-4. Conceptual model showing the relationships among ambient air quality**  
 3 **indicators and exposure pathways and the resulting impacts on ecosystems,**  
 4 **ecological responses, effects and benefits to characterize known or**  
 5 **anticipated adverse effects to public welfare.**  
 6

7 These concepts can also be applied to the programs described in section 3.1. National  
 8 parks represent areas of nationally recognized ecological and public welfare significance, which  
 9 are afforded a higher level of protection. Therefore, staff has also focused on air quality and  
 10 deposition in the subset of national park sites and important natural areas. The spatial  
 11 relationships between sensitive regions, Class 1 areas and nitrogen deposition levels are  
 12 illustrated in Figures 3-5 and 3-6. Please note that the scale of deposition levels is different for  
 13 the two maps to allow greater differentiation of the deposition in the western U.S.



1  
 2 **Figure 3-5. Locations of Eastern U.S. Public Lands relative to deposition of nitrogen and**  
 3 **sulfur in sensitive aquatic areas. Source 2005 CMAQ and NADP.**  
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**Figure 3-6. Location of Western U.S. Public Lands relative to deposition of nitrogen and sulfur. Source 2005 CMAQ and NADP.**

### 3.3 Applying Economic Valuation to Ecosystem Services

As discussed earlier in this document, a secondary NAAQS is required to be set at the “level(s) of air quality necessary to protect the public welfare from any known or anticipated adverse effects”. As part of the effort to determine the standard, EPA linked the changes in the ambient air concentrations of NO<sub>y</sub> and SO<sub>x</sub> to the changes in ecosystem services and ultimately to changes in public welfare (U.S. EPA, 2009). The difficulty in the monetization for ecosystem services has been previously emphasized. This difficulty necessitates focusing on a subset of services for economic valuation. And although economics on its own cannot determine what level of impact on public welfare is “adverse,” economics can be helpful in the context of a secondary NAAQS for determining the degree to which improvements are beneficial to public welfare and illustrating and aggregating those impacts.<sup>1</sup>

#### 3.3.1 Ecosystem Services and Links to Public Welfare

An ecosystem service framework provides a structure to measure changes in public welfare from changes in ecosystem functions affected by air pollution. EPA’s Risk Assessment for this rulemaking defines ecosystem services as “the ecological processes or functions having monetary or nonmonetary value to individuals or society at large” (EPA 2009). The discipline of economics provides a useful approach for summarizing how the public values changes in the services provided by the environment. An ecosystem services framework (with or without valuation) can provide measures of changes in public welfare.

#### 3.3.2 Economics as a Framework to Illustrate Changes in Public Welfare

Economics can provide a framework to illustrate how public welfare changes in response to changes in environmental quality by quantitatively linking changes in ecosystem services to preferences. Economics assumes that the choices that individuals make reflect their preferences over certain outcomes and that, generally speaking, they will make choices that, in expectation, will make them as well off as possible given their resources. In economics revealed and stated

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<sup>1</sup> Section 109 of the Clean Air Act forbids consideration of the compliance costs of reducing pollution when setting a NAAQS. However, there is no prohibition regarding the consideration of the monetized impacts of welfare effects occurring due to levels of pollution above alternative standards in evaluating the adversity of the impacts to public welfare. Ecosystem services can be characterized as a method of monetizing the impacts of the air pollution. Although a separate regulatory document quantifying the costs and benefits of attaining a NAAQS is prepared simultaneously, this document is not considered when selecting a standard.

1 preference methods are used to observe the choices individuals make to understand the outcomes  
2 individuals prefer. What individuals are willing to give up for an outcome is their willingness-to-  
3 pay (WTP) for that outcome. An example of an outcome is an improvement in an ecosystem  
4 service. Often, to provide comparability to other goods and services, in economics these  
5 tradeoffs are framed relative to dollars for convenience.

6 Economics could inform the Administrator by valuing and characterizing the changes in  
7 public welfare from changes in the quantity and quality of ecosystem services. Overall, this  
8 assessment intends to characterize changes in ecosystem services from a scientific perspective  
9 using effects on ecosystem structures and functions or ecosystem integrity. Economics then  
10 estimates the effect on public welfare of these changes in the quantity and quality of ecosystem  
11 services using willingness to pay as a measure of this effect. For example, a decrease in a  
12 particular bird species can be characterized by its effect on the ecosystem's structure and  
13 function, while from an economic perspective, the effects would be based on the impact on  
14 public welfare or the value the public places on that species. A simple example is a comparison  
15 between a decrease in a bird species that is relatively unknown compared to a decrease in a very  
16 prominent species (e.g. bald eagle). The public is likely to have a higher WTP to avoid the latter,  
17 and thus the decrease would affect the public welfare more, even if the changes in the two bird  
18 species generally have the same impact on an ecosystem's structure or function.

19 There are important complications with using preferences to understand the effect of  
20 pollution on public welfare. For example, while the field of economics generally assumes that  
21 public preferences are the paramount consideration; care must be taken that these preferences  
22 may change when the public receives new information. Evaluation of public preferences should  
23 take place under conditions of full information. If individuals do not understand how pollution  
24 will affect ecosystem services, or even how those ecosystem services affect their quality of life,  
25 then they will have a difficult time valuing changes in those services. Similarly, it may be very  
26 difficult and time-consuming for individuals to learn and understand how changes in particular  
27 ecosystem services may affect them, in part because typically there are significant  
28 interdependencies within an ecosystem. Because of this complexity, individuals may implicitly  
29 value a species, or habitat, or ecosystem function because it supports an ecosystem service that  
30 they do clearly value. Furthermore, the public also has limited understanding regarding  
31 irreversibilities, tipping points, and other more complex aspects of ecosystems, which limits the

1 ability to adequately value these ecosystems.<sup>2</sup> In addition, where and when a change in an  
2 ecosystem takes place is crucial for characterizing the associated change in an ecosystem  
3 service, and will also affect the value the public places on that change.

### 4 5 **3.3.3 The Role of Economics in Defining “Adversity”**

6 If economic valuation can establish a significant loss to public welfare, then this can  
7 provide strong support for a determination of an “adverse” effect. However, there is neither an  
8 economic definition of how much loss in public welfare is adverse nor an economic definition of  
9 adversity. While an economist might consider a particular scenario adverse because it might  
10 imply some harm or potential for improvement, there is no specific threshold level when a loss in  
11 welfare (e.g. loss in dollars) becomes adverse. An individual might be willing to give up some  
12 of their resources to avoid a threat or negative outcome (i.e., willing to pay to avoid a particular  
13 outcome). According to economic theory, if an individual is willing to give up something to  
14 avoid the outcome, then imposing the outcome on the individual must make them worse off, at  
15 which point an economist might describe the outcome as adverse. However, the amount an  
16 individual is willing to pay to avoid the outcome may or may not rise to the level of harm that the  
17 Administrator interprets as “adverse” to public welfare. At the same time, an economic  
18 valuation that shows that there are substantial damages from current levels of acidification or  
19 nutrient enrichment would provide strong evidence for finding that current impacts are adverse  
20 to public welfare. In summary, economic analysis (particularly valuation) can provide useful  
21 information for the Administrator as to the interpretation of the word “adverse” in the context of  
22 public welfare, but it does not provide a complete set of information needed to make that  
23 determination.

### 24 25 **3.3.44 Collective Action as an Indicator of Public Preferences**

26 Typically economics uses information on willingness to pay for improved environmental  
27 quality that is gathered from observing individuals’ market behavior (revealed preference) or that  
28 they provide through surveys (i.e., stated preference methods). The analyses in the following

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<sup>2</sup> While the public may not fully appreciate the interdependencies within ecosystems, they can learn them, but again it may be costly to do so. It is possible for individuals to value outcomes that are irreversible or result in discrete changes (i.e., tipping points) in the quality and quantity of ecosystem services. Avoiding irreversible outcomes should be and are more valued by individuals than outcomes that are not irreversible (Arrow and Fischer, 1974).

1 sections use revealed and stated-preference information to quantify a portion of the social costs  
2 of current levels of acidification and nutrient enrichment. However, the studies supporting these  
3 analyses evoke specific contexts and thus the findings may not be generalizable across all of  
4 those affected by acidification or nutrient enrichment.<sup>3</sup> An alternative source of revealed  
5 information on the damages caused by acidification can be found in the behavior of groups.  
6 Often groups collectively make choices to engage in activities that improve the collective  
7 welfare of the group. For example, a community around an acidified lake might undertake  
8 activities designed to improve the quality of that lake, including purchasing lime, to use as a tool  
9 to reduce the acidity of the lake. These collective decisions can be used to gain insights into how  
10 people value improvements to ecosystem services. However, there are many obstacles to  
11 collective actions, including problems of organization, free ridership and others (Olson 1965)  
12 that make it difficult to use the actions of organizations to interpret individuals' preferences.

13 In addition to communities, states may also take actions to increase the quality of their  
14 impaired lakes. Non-Governmental Organizations (NGOs) or advocacy groups, as well may  
15 organize support for, and/or directly undertake, activities to improve lake and stream quality on  
16 behalf of its members/donors. How individual's preferences are expressed through these  
17 collective actions is discussed below. For brevity, this discussion will focus on collective efforts  
18 to reduce acidity of lakes and streams by Communities, Nongovernmental Organizations and  
19 States.

#### 20 21 *3.3.4.1 Communities*

22 In cases where property rights to a resource are well defined, collective action is more  
23 likely to take place, as individuals have greater ownership and control over the affected resource.  
24 Rights to use a lake, as well as, mandatory membership in a lake association is often written right  
25 into the deed of properties which abut or surround a lake, giving these property owners more  
26 control over the resource. This mechanism of granting rights and responsibilities to the lake  
27 encourages better management of the lake resource by remedying unrestricted access and free  
28 rider problems. This coupling of the costs of resource improvements with their benefits  
29 encourages individuals to maintain the quality of the resource.

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<sup>3</sup> Even in the case where the existing studies provide a reliable characterization of the effects of acidification or nutrient enrichment on a limited number of individuals, it is advisable to make use of corroborating data and studies when such information is available.

1           There have been several documented instances where communities (particularly  
2 Homeowners Associations) have spent time and money to improve the quality of a lake. These  
3 include actions to combat acidity, eutrophication, invasive species (e.g. Zebra Mussels) and other  
4 problems. The Lake Wononscopomuc Association in Salisbury, Connecticut is a typical  
5 example (Mayland 2009.) They spend their own funds to hire scientific consultants to survey  
6 and test the lake water (for e.g. pH., dissolved oxygen, visibility, and many other factors related  
7 to the lake's condition) and recommend management strategies to improve the quality of the  
8 lake. Likewise, in Georgia, the Berkeley Lake Homeowners Association (BLHA) is a non-profit  
9 homeowner association dedicated to protecting Berkeley Lake. BLHA is typical of many other  
10 home owners associations with access to a lake, in that they are also concerned with and  
11 managing acidity, eutrophication, invasive species and a whole host of more mundane upkeep  
12 and maintenance issues (Hunkapiller 2006.) BLHA recognizes the relationship between lake  
13 acidity and resident's enjoyment of the lake's fishing swimming and aesthetics.

14

#### 15           3.3.4.2 *Nongovernmental Organizations*

16           Nongovernmental Organizations (NGOs) or Advocacy Groups organize individuals and  
17 smaller groups thereby reducing the transaction costs associated with individual's desires to  
18 advance a specific goal. For example, Living Lakes, Inc. (LLI) is a not-for profit organization  
19 which sponsors an applied aquatic resources restoration demonstration program for acidified  
20 waters. In the late 1980's LLI began evaluation of seven different liming technologies on 22  
21 lakes and 10 streams in 6 states. Lakes and streams have been treated in the states of  
22 Massachusetts, Rhode Island, New York, Pennsylvania, Maryland and West Virginia (Brocksen  
23 and Emler 1988.) Likewise, sportsman groups such as Trout Unlimited, as well as, smaller local  
24 groups, have an interest in improving or maintaining the quality of lakes and streams. Trout  
25 Unlimited is well known for these activities and is discussed further later on. However, several  
26 smaller, localized groups also work to decrease aquatic acidification. One of these is the  
27 Mosquito Creek Sportsman's Association in Pennsylvania. Mosquito Creek and its main  
28 tributary Gifford Run were once famous for naturally reproducing wild brook and brown trout.  
29 However, since the early 1960's, the pH of the stream steadily declined due to acid rain. As a  
30 result, wild brook trout, as well as, wild brown trout have substantially declined in the watershed  
31 (Hoover and Rightnour 2002.) Aerial liming was undertaken as part of an overall watershed  
32 restoration program that included constructed wetlands, forest liming, and in-stream liming to

1 improve this fishery and provide increased opportunities for public recreation in the region.  
2 Fifty tons of lime were applied in the headwaters of Mosquito Creek Watershed. This liming  
3 project was part of the Mosquito Creek Sportsman’s Association’s efforts to improve the water  
4 quality of the 90 square mile watershed located in Clearfield and Elk counties. However, while  
5 the project first phase and the other ongoing phases of the overall restoration project have been  
6 initiated by the Mosquito Creek Sportsman Association, they received technical support from  
7 multiple public, private and other non-profit groups.<sup>4</sup> “A benefit/cost analysis was prepared on  
8 the four implementation phases of this project. Costs were based on alkaline deficiencies and the  
9 additional costs determined for the technologies. Benefits were estimated as returns on direct  
10 recreational use losses and community willingness-to-pay. Full restoration of the watershed is  
11 estimated to cost approximately \$3.4 million over 15 years, for an annualized cost of \$229,000,  
12 or \$5,400 per mile per year for 42 miles of potential improvements. Expected returns range from  
13 \$1.2 million per year for recreational use to \$6.1 million per year for total community  
14 willingness-to-pay. It was concluded that restoration is technically feasible and economically  
15 beneficial for the Mosquito Creek watershed, and it is recommended that planned projects and  
16 the remainder of the progressive restoration plan be implemented.” (Hoover and Rightnour  
17 2002.)

18

### 19 3.3.4.3 States

20 Several states including: Vermont, New Hampshire, New York and Tennessee have  
21 developed Total Maximum Daily Loads (TMDLs) for lakes impaired for acidification in their  
22 jurisdictions. As mentioned in the previous section regarding the TMDL for the Chesapeake Bay  
23 the applicable water quality standards and designated uses are set by the states with public  
24 participation. Although most states set their standard either by legislation or regulations in at  
25 least one case, specifically New York, the designated uses and water quality standard are part of  
26 the state constitution. The Adirondack Forest Preserve is required to be “forever kept as wild  
27 forest lands”. New York has interpreted this to mean that the waters included in the preserve are

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<sup>4</sup> These included: the Pennsylvania State University Environmental Resource Research Institute, Pennsylvania Game Commission, Pennsylvania DCNR Bureau of Forestry, Pennsylvania Fish and Boat Commission, Pennsylvania Department of Environmental Protection, Pennsylvania Department of Corrections Quehanna Boot Camp, Wood Duck Chapter Trout Unlimited, Canaan Valley Institute, Clearfield County Conservation District and Pennsylvania USDA Natural Resource Conservation Service.

1 required to be kept in natural conditions. To this end New York has chosen to set a tiered TMDL  
2 that allows interim water quality targets in order to address the reality that some lakes in the  
3 Adirondacks will naturally have a pH that does not meet the state's water quality standards. For  
4 lakes that can meet the standards the state has chosen to set the water quality standard for pH is  
5 above 6.5. New Hampshire has chosen to set their water quality target at an ANC of 60µeq/L  
6 that, according to the TMDL document, corresponds to a pH of 6.5. Vermont, in a similar  
7 process has chosen a target ANC of 50µeq/L. In Tennessee the state faces a similar problem as  
8 New York in trying to set levels to protect streams Great Smoky Mountains National Park which  
9 include some naturally acidic streams. Accordingly they have set site specific ANC targets  
10 where data is available to do so and chosen to target an ANC of 50µeq/L as a default value  
11 where data is not available. The Tennessee TMDL is a partnership between the state and the  
12 National Park Service which is sharing the data collection and modeling activities with academic  
13 institutions.

14 In each case the implementation sections of these TMDLs cites the fact that the sources  
15 of pollution responsible for the degradation of water quality in the named lakes and streams are  
16 not located within the jurisdiction of the state. Each state has called on EPA to require regional  
17 or national decreases in acidifying deposition. Vermont goes so far as to say "In short,  
18 implementation of this TMDL is primarily the responsibility of EPA.... This TMDL sets out  
19 clear endpoints to guide EPA actions. However, in the absence of vigorous efforts by EPA to  
20 bring about reductions in acid emissions from out-of-state sources this TMDL will merely have  
21 been a paper exercise."

22

#### 23 *3.3.4.4 Public-Private Collaborations*

24 In some cases, industry, government and private efforts partner to reduce the acidity of a  
25 lake or stream. In one such instance in 2005, The U.S. Forest Service used helicopters to apply  
26 200 tons of limestone sand into the St. Marys River and its tributaries to lower acid levels in one  
27 of Virginia's prime trout fisheries to mitigate the impacts of acidification until a long-term  
28 solution to acidification is found. The NGO, Trout Unlimited was one of the partners in the  
29 liming project, while Dominion Virginia Power provided \$10,000 for the liming project  
30 (Associated Press 2005.) In another partnership, Living Lakes participated in a project in the  
31 Woods Lake Watershed in the Adirondack region of the state of New York that was co-

1 sponsored by the Electric Power Research Institute (EPRI.) There are many such examples,  
2 where all three of these types of groups partner on the same project.

#### 3 4 *3.3.4.5 Using Information on Collective Actions to Estimate Welfare Impacts*

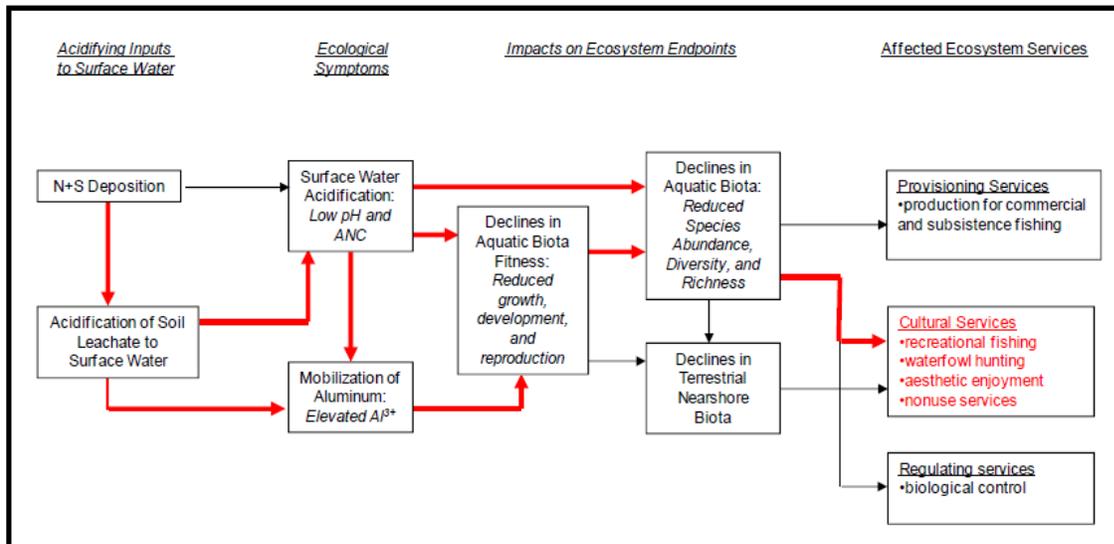
5 The fact that collective action activities are being undertaken by communities, NGO's  
6 and States underscores the fact that there is a societal demand for further improvement to the  
7 quality of many water bodies which have been impaired by acidic deposition.<sup>5</sup> However, they  
8 provide insufficient quantitative evidence as to what society willingness to pay to reduce lake  
9 and stream acidity because it is difficult to separately identify individual preferences from the  
10 actions of the group. For example, groups suffer from the problem of free-ridership. When there  
11 are free-riders we know that the activity of the group understates the preferences of the  
12 community for an improvement in environmental quality, however it is difficult to estimate the  
13 magnitude of its impact on the total activity of the group.

### 14 15 **3.4 Effects of Acidification and Nutrient Enrichment on Ecosystem Services**

16 The process used to link ecological indicators to ecosystem services is discussed  
17 extensively in Appendix 8 of the REA. In brief, for each case study area assessed the ecological  
18 indicators were linked to an ecological response that was subsequently linked, to the extent  
19 possible, to associated services. For example in the case study for aquatic acidification the  
20 chosen ecological indicator is ANC which can be linked to the ecosystem service of recreational  
21 fishing as illustrated in the conceptual model shown in Figure 3-7. Although recreational fishing  
22 losses are the only service effects that can be independently quantified or monetized at this time,  
23 there are numerous other ecosystem services that may be related to the ecological effects of  
24 acidification.

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<sup>5</sup> However, one must recognize that often times reducing acidity is often part of a larger effort to generally improve the quality of a water body. Therefore, separating out the portion of people's desire to just to reduce acidity from the more general improvements is difficult.



1  
 2 **Figure 3-7. Conceptual model linking ecological indicator (ANC) to affected ecosystem**  
 3 **services. The red arrows highlight the path to monetization of recreational**  
 4 **fishing effects.**

5  
 6 While aquatic acidification is the focus of this policy assessment, the other effect areas  
 7 analyzed in the REA still merit some discussion in view of the fact that these ecosystems are  
 8 being harmed by nitrogen and sulfur deposition and will obtain some measure of protection with  
 9 any decrease in that deposition regardless of the reason for the decrease. In next four sections we  
 10 summarize the current levels of specific ecosystem services for aquatic and terrestrial  
 11 acidification, and aquatic and terrestrial nutrient enrichment. We also present results of analyses  
 12 that have attempted to quantify and monetize the harms to public welfare, as represented by  
 13 ecosystem services, due to nitrogen and sulfur deposition.

14 For the purposes of the following sections nutrient enrichment refers only to that due to  
 15 NO<sub>y</sub> deposition. Additionally these sections focus on the detrimental effects of that deposition.  
 16 Staff acknowledges that a certain amount of NO<sub>y</sub> deposition in managed terrestrial ecosystems  
 17 may have a beneficial effect, specifically increased growth (a fertilization effect). However no  
 18 attempt has been made to quantify those beneficial effects since this document and preceding  
 19 analyses are focused on unmanaged sensitive ecosystems.

20  
 21  
 22

### 3.4.1 Evidence for Adversity Related to Aquatic Acidification

Acidification primarily affects the ecosystem services that are derived from the fish and other aquatic life found in these surface waters (**REA, Section 5.2.1.3**). In the northeastern United States, the surface waters affected by acidification are not a major source of commercially raised or caught fish; however, they are a source of food for some recreational and subsistence fishers and for other consumers. Although data and models are available for examining the effects on recreational fishing, relatively little data are available for measuring the effects on subsistence and other consumers. For example, although there is evidence that certain population subgroups in the northeastern United States, such as the Hmong and Chippewa ethnic groups, have particularly high rates of self-caught fish consumption (Hutchison and Kraft, 1994; Peterson et al., 1994), it is not known if and how their consumption patterns are affected by the reductions in available fish populations caused by surface water acidification.

Inland surface waters support several cultural services, such as recreational fishing, aesthetic and educational services; however, Banzhaf et al (2006) has shown that non-use services are arguably a significant source of benefits from reduced acidification. The areas of the country containing the most sensitive lakes and streams are New England, the Adirondack Mountains, the Appalachian Mountains (northern Appalachian Plateau and Ridge/Blue Ridge region), northern Florida, and the Upper Midwest. Within the Adirondack Mountains approximately 8% of the lakes were considered acidic and in the northern Appalachian Plateau and Ridge/Blue Ridge 6 – 8% of the streams (**ISA 3.2.4.2 and REA 4.2.2**). Recreational fishing in lakes and streams is among the most popular outdoor recreational activities in the northeastern United States. Data from the 2006 National Survey of Fishing, Hunting, and Wildlife Associated Recreation (FHWAR) indicate that more than 9% of adults in this part of the country participate annually in freshwater fishing with 140 million freshwater fishing days. Based on studies conducted in the northeastern United States, Kaval and Loomis (2003) estimated average consumer surplus values per day of \$35 for recreational fishing (in 2007 dollars). Therefore, the implied total annual value of freshwater fishing in the northeastern United States was \$5 billion in 2006. We recognize that embedded in these numbers is a degree of harm to recreational fishing services due to acidification that has occurred over time. These harms have not been quantified on a regional scale. However given the magnitude of the resource, the length of time nitrogen and sulfur have been affecting freshwaters in the northeast and the level of monetary

1 damages calculated for the case study in the Adirondacks described in the next section we would  
2 expect these damages to be significant.

3 In general, inland surface waters such as lakes, rivers, and streams provide a number of  
4 regulating services, playing a role in hydrological regimes and climate regulation. There is little  
5 evidence that acidification of freshwaters in the northeastern United States has significantly  
6 degraded these specific services; however, freshwater ecosystems also provide biological control  
7 services by providing environments that sustain delicate aquatic food chains.

8 The toxic effects of acidification on fish and other aquatic life impair these services by  
9 disrupting the trophic structure of surface waters (Driscoll et al., 2001). Although it is difficult  
10 to quantify these services and how they are affected by acidification, it is worth noting that some  
11 of these services may be captured through measures of provisioning and cultural services. For  
12 example, these biological control services may serve as “intermediate” inputs that support the  
13 production of “final” recreational fishing and other cultural services.

14

#### 15 **3.4.1.1 What is the value of the impaired recreational fishing and other cultural services?**

16 In the previous section we described the ecosystem services that are most likely to be  
17 affected by N and S deposition and summarized evidence regarding the current magnitude and  
18 values of recreational fishing services, the degree to which these services are impaired by  
19 existing NOy/SOx levels has not been quantified. To address this limitation, the **REA**  
20 (**Appendix 8**) provides insights into the magnitude of ecosystem service impairments. The  
21 REA provides quantitative estimates of selected ecosystem services impairments or  
22 enhancements for three main categories of ecosystem effects – aquatic acidification, terrestrial  
23 acidification, and aquatic nutrient enrichment<sup>6</sup>. Within these three categories, the selection of  
24 specific ecosystem services for more in-depth analysis depended primarily on the expected  
25 magnitude of impairments and on the availability of appropriate data and modeling tools.

26 The analysis of ecosystem service impairments due to aquatic acidification builds on the  
27 case study analysis of lakes in the New York Adirondacks. In this study estimates of changes in  
28 recreational fishing services are determined, as well as changes more broadly in “cultural”  
29 ecosystem services (including recreational, aesthetic, and nonuse services). First, the MAGIC

---

<sup>6</sup> Estimates for terrestrial nutrient enrichments were not generated due to the limited availability of necessary data and models for this effect category.

1 model (REA, Appendix 8, Sec 2.2) was applied to 44 lakes to predict what ANC levels would  
 2 be under both “business as usual” conditions (i.e., allowing for some decline in deposition due to  
 3 existing regulations) and pre-emission (i.e., background) conditions. These model runs assumed  
 4 a 2010 “zero-out” emissions scenario (where all N and S deposition is eliminated) with a  
 5 projected lag time between the elimination of emissions to observed improvement in ANC of 10  
 6 years thus benefits results were calculated for the year 2020. These predictions were then  
 7 extrapolated to the full universe of Adirondack lakes. Table 3-2 reports the number of  
 8 “impacted” lakes in each year, where impact means that the lake is predicted to be below the  
 9 ANC threshold under business-as-usual *and* above the threshold under pristine conditions.

10  
11

**Table 3-2. Count of Impacted Lakes**

<b>ANC Threshold (in µeq/L)</b>	<b>Year</b>	<b>Lake Count</b>
20	2005	0
20	2020	107
20	2050	95
20	2100	74
50	2005	0
50	2020	244
50	2050	222
50	2100	200
100	2005	0
100	2020	430
100	2050	404
100	2100	354

12 Note: There are 1,076 lakes in the “Adirondack Region”.

13 Second, to estimate the recreational fishing impacts of aquatic acidification in these lakes,  
 14 an existing model of recreational fishing demand and site choice was applied. This model  
 15 predicts how recreational fishing patterns in the Adirondacks would differ and how much higher  
 16 the average annual value of recreational fishing services would be for New York residents if lake  
 17 ANC levels corresponded to background (rather than business as usual) conditions. Table 3 – 3

1 summarizes the results and the present value of benefits and annualized benefits at 3 and 7%  
 2 discount rates.

**Table 3-3. Present Value and Annualized Benefits, Adirondack Region**

ANC Threshold (in µeq/L)	Present Value Benefits <sup>a</sup> (in million of 2007 dollars)		Annualized Benefits <sup>b</sup> (in million of 2007 dollars)	
	3% Discount Rate	7% Discount Rate	3% Discount Rate	7% Discount Rate
20	\$142.59	\$60.05	\$4.46	\$3.94
50	\$285.15	\$114.18	\$8.91	\$7.49
100	\$298.67	\$120.61	\$9.33	\$7.91

3 <sup>a</sup> Annual benefits for 2010 to 2100 discounted to 2010.

4 <sup>b</sup> Present value benefits annualized over 2009–2100.

5 Current annual impairments are most likely of a similar magnitude because, although  
 6 current NOy/SOx levels are somewhat higher than those expected in 2020 (under business as  
 7 usual – given expected emissions controls associated with Title IV regulations but no additional  
 8 nitrogen or sulfur controls), and the affected NY population is also somewhat smaller (based on  
 9 U.S. Census Bureau projections).

10 To estimate impacts on a broader category of cultural (and some provisioning)  
 11 ecosystem services, results from the Banzhaf et al (2006) valuation survey of New York  
 12 residents were adapted and applied to this context. The survey used a contingent valuation  
 13 approach to estimate the average annual household WTP for future reductions (20% and 45%) in  
 14 the percent of Adirondack lakes impaired by acidification. The focus of the survey was on  
 15 impacts on aquatic resources. Pretesting of the survey indicated that respondents nonetheless  
 16 tended to assume that benefits would occur in the condition of birds and forests as well as in  
 17 recreational fishing. The survey that measured the benefits of 20% of the lakes improving  
 18 indicated that terrestrial benefits were minor and econometric controls were used to adjust the  
 19 willingness to pay estimate for those that suspected that terrestrial improvements were greater  
 20 than described in the survey. The survey that measured the benefits of improving 45% of the  
 21 total number of lakes also indicated that the benefits to forests and birds were significant. .

22 The WTP estimates from the two versions of the survey were then (1) scaled to reflect  
 23 predicted changes between business-as-usual and background conditions in 2020 (MAGIC lake

1 modeling results indicate that impaired lakes would decrease from 22 to 31% using background  
 2 conditions with ANC increasing from 20 to 50 $\mu$ eq/L), and (2) aggregated across New York  
 3 households. The scaling entails converting the average household willingness-to-pay for the  
 4 improvements described in the Adirondacks surveys to an average household willingness-to-pay  
 5 per percentage point of the total population of lakes improved.<sup>7</sup> The results are summarized in  
 6 Table 3-4. The range of average household willingness to pay reflects the range in willingness to  
 7 pay per percentage point of lakes improved described in the two versions of the survey.  
 8 Estimates are provided at ANC 20, 50, and 100 to reflect the range of ANC discussed  
 9 throughout the REA and this document and for consistency with the Random Utility Model  
 10 analysis.

**Table 3-4. Aggregate Annual Benefit Estimates for the Zero-Out Scenario**

ANC Threshold	Reduction in Percentage of Unhealthy Lakes	Range of Average Household WTP per Percentage Reduction		Number of NY Households (in millions)	Range of Annual Benefits (in millions of 2007 \$)	
20 $\mu$ eq/L	22%	\$2.63	\$5.87	7.162	\$410.6	\$916.4
50 $\mu$ eq/L	31%	\$1.32	\$3.76	7.162	\$291.2	\$829.4
100 $\mu$ eq/L	26%	\$2.63	\$5.87	7.162	\$491.6	\$1,097.2

11  
 12 These results suggest that the value of avoiding current impairments to ecosystem  
 13 services from Adirondack lakes are even higher than the estimate, because the estimates assume  
 14 a lag of 10 years in which no benefits accrue and because the percent of impaired lakes is slightly  
 15 higher today than expected in 2020 under business-as-usual. These results imply significant  
 16 value to the public in addition to those derived from recreational fishing services. Note that the  
 17 results are only applicable to improvements in the Adirondacks valued by residents of New  
 18 York. If similar benefits exist in other acid-impacted areas, benefits for the nation as a whole  
 19 could be substantial. The analysis provides results on only a subset of the impacts of acidification  
 20 on ecosystem services and suggests that the overall impact on these services is likely to be  
 21 substantial.

22

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<sup>7</sup> Scaling is required because neither of the surveys administered by Banzhaf et al. (2006) describe improvements that correspond exactly to the improvement scenario modeled here.

### 3.4.2 Evidence for Adversity Related to Terrestrial Acidification

In the previous chapter of this document we discussed the effects of acidifying deposition on terrestrial ecosystems, especially forests. These include the observed decline and/or dieback in red spruce and sugar maple. These species are particularly sensitive to acidifying deposition and have ranges that overlap the areas of the U.S. where some of the highest levels of acidifying deposition occur. Additionally these species are present in the case study areas examined in the REA. As a result we chose to focus on red spruce and sugar maple as the species of interest for the analysis of ecosystem services presented in this section.

A similar model to Figure 3-6 can be drawn for terrestrial acidification that links Bc:Al molar ratio to reduced tree growth to decreases in timber harvest, although we have less confidence in the significance of this linkage than we do for aquatic acidification. There are numerous services expected to be affected but the data and methods to adequately describe those losses does not as yet exist. These services include effects to forest health, water quality, and habitat, including decline in habitat for threatened and endangered species, decline in forest aesthetics, decline in forest productivity, increases in forest soil erosion and decreases in water retention (ISA, 2009; REA, 2009; Krieger, 2001). Forests in the northeastern United States provide several important and valuable provisioning services, which are reflected in the production and sales of tree products.

Sugar maples are a particularly important commercial hardwood tree species in the United States, producing timber and maple syrup that provide hundreds of millions of dollars in economic value annually (NASS, 2008). Red spruce is also used in a variety of wood products and provides up to \$100 million in economic value annually (USFS, 2006).

Forests in the northeastern United States are also an important source of cultural ecosystem services, including nonuse (existence value for threatened and endangered species), recreational, and aesthetic services (ISA, 2009; REA, 2009). Red spruce forests are home to two federally listed species, the spruce-fir moss spider and the rock gnome lichen. The value of these two endangered species has not been estimated.

Although we do not have the data to link acidification damages directly to economic values of lost recreational services in forests, these resources are valuable to the public. For example, most recent data from the National Survey on Recreation and the Environment (NSRE) indicate that, from 2004 to 2007, 31% of the U.S. adult (16 and older) population visited a

1 wilderness or primitive area during the previous year, and 32% engaged in day hiking (Cordell et  
 2 al., n.d.). A recent study suggests that the total annual value of off-road driving recreation was  
 3 more than \$9 billion, total and value of hunting and wildlife viewing was more than \$4 billion  
 4 each in the Northeastern United States in 2006 (Kaval and Loomis, 2003). Table 3-5  
 5 summarizes data from the NSRE and the Fishing, Hunting, and Wildlife Related Activity Survey  
 6 (U.S. DOI, 2007) along with average WTP estimates from Kaval and Loomis to estimate the  
 7 total value of these services in the northeast.

8  
 9 **Table 3-5. Annual participation and value of outdoor (forest related)**  
 10 **activity in the northeast.**

<b>Recreational Activity</b>	<b>Participation Rate (%)</b>	<b>Activity Days (in Thousands)</b>	<b>Avg. WTP Per Activity Day (\$2007)</b>	<b>Total Value (in millions)</b>
Off Road Driving	16	366,336	\$25.25	\$9,250
Hunting	5.5	83,821	\$52.36	\$4,380
Wildlife Viewing	10	122,200	\$34.46	\$4,210

11  
 12 In addition, fall color viewing is a recreational activity that is directly dependent on  
 13 forest conditions. Sugar maple trees, in particular, are known for their bright colors and are,  
 14 therefore, an essential aesthetic component of most fall color landscapes. Statistics on fall color  
 15 viewing are much less available than for the other recreational and tourism activities; however, a  
 16 few studies have documented the extent and significance of this activity. For example, Spencer  
 17 and Holecek (2007) found that approximately 30% of residents in the Great Lakes area reported  
 18 at least one trip in the previous year involving fall color viewing. In a separate study conducted  
 19 in Vermont, Brown (2002) reported that more than 22% of households visiting Vermont in 2001  
 20 made the trip primarily for the purpose of viewing fall colors.

1 Two studies have estimated values for protecting high-elevation spruce forests in the  
 2 Southern Appalachians. Kramer et al. (2003) conducted a contingent valuation study estimating  
 3 households' WTP for programs to protect remaining high-elevation spruce forests from damages  
 4 associated with air pollution and insect infestation (Haeefele et al., 1991; Holmes and Kramer,  
 5 1995). The survey presented respondents with a sheet of color photographs representing three  
 6 stages of forest decline and explained that, without forest protection programs, high-elevation  
 7 spruce forests would all decline to worst conditions (with severe tree mortality) and two potential  
 8 forest protection programs. Median household WTP was estimated to be roughly \$29 (in 2007  
 9 dollars) for the minimal program and \$44 for the more extensive program. Another study by  
 10 Jenkins, Sullivan, and Amacher (2002) estimated an aggregate annual value of \$3.4 billion for  
 11 avoiding a significant decline in the health of high-elevation spruce forests in the Southern  
 12 Appalachian region.

13  
 14 **Table 3-6. Summary of Studies of Select Terrestrial Ecosystem Services**

Fall Color Viewing	30%	Great Lakes area residents	Spencer (2007)
	22%	Vermont visitors	Brown (2002)
Protection of spruce		Southern Appalachians	
	\$29	WTP per household for minimal program	Kramer et al. (2003)
	\$44	WTP per household for extensive program	
	\$3.4 b	Aggregate annual value	Jenkins (2002)

15  
 16 Forests in the northeastern United States also support and provide a wide variety of  
 17 valuable regulating services, including soil stabilization and erosion control, water regulation,  
 18 and climate regulation (Krieger, 2001). Forest vegetation plays an important role in maintaining  
 19 soils in order to reduce erosion, runoff, and sedimentation that can adversely impact surface  
 20 waters. In addition to protecting the quality of water in this way, forests also help store and help  
 21 regulate the quantities and temporal discharge patterns of water in watersheds. Forests also play

1 an important role in carbon sequestration at both regional and global scales. The total value of  
2 these ecosystem services is very difficult to quantify.

3 **3.4.2.1 What is the value of current ecosystem service impairments?** The REA  
4 Appendix 8 describes an analysis of ecosystem service impairments associated with the impacts  
5 of terrestrial acidification on the forest product provisioning services from two commercially  
6 important tree species on unmanaged forests – sugar maple and red spruce - that are particularly  
7 sensitive to the effects of acidification. Evidence of effects due to terrestrial acidification is  
8 particularly strong for these two species whose range includes the northeastern U.S. where levels  
9 of nitrogen and sulfur deposition have historically been relatively high, however more  
10 widespread impacts that include other tree species are also possible. We acknowledge that there  
11 may be some beneficial fertilization effects of nitrogen deposition however given the complexity  
12 of the nitrogen cycle it is not possible to quantify all those effects here. There is a detailed  
13 discussion of nitrogen fertilization effects in Chapter 4.

14 In an exploratory study that is still under development we used data from the USFS  
15 Forest Inventory and Analysis (FIA) database, to estimate an exposure-response relationship for  
16 each species to measure the average negative effect of critical load exceedances (CLEs) of  
17 nitrogen and sulfur deposition on annual tree growth. These estimated relationships were then  
18 applied to sugar maple and red spruce stocks in the Northeast and North central regions to  
19 estimate the average percent increase in annual tree growth that would occur if all CLEs were  
20 eliminated. To estimate the aggregate-level forest market impacts of eliminating CLEs starting  
21 in the year 2000, the tree-level growth adjustments were applied using the Forest and  
22 Agricultural Sector Optimization Model (FASOM), which is a dynamic optimization model of  
23 the U.S. forest and agricultural sectors. The model results are reported as the present discounted  
24 values of future welfare changes in the forestry sector (in 5-year increments from 2000 to 2080)  
25 due to increased tree growth. Summing over this 80-year period, the total present value of these  
26 welfare gains is \$40.705 million (in 2006 dollars, using a 4% discount rate). On an annualized  
27 basis (at 4%), this is equivalent to \$1.64 million per year. These estimates can also be  
28 interpreted as the current value of impairments to forest provisioning services provided by red  
29 spruce and sugar maple due to acidification effects from nitrogen and sulfur. These results  
30 should be considered very uncertain due to the pending revision of the exposure – response curve  
31 and release of an updated version of the FASOM model.

### 3.4.3 Evidence for Adversity Related to Aquatic Nutrient Enrichment

Estuaries in the eastern United States are important for fish and shellfish production. The estuaries are capable of supporting large stocks of resident commercial species, and they serve as the breeding grounds and interim habitat for several migratory species (U.S. EPA, 2009). To provide an indication of the magnitude of provisioning services associated with coastal fisheries, from 2005 to 2007, the average value of total catch was \$1.5 billion per year in 15 East Coast states. It is not known, however, what percentage of this value is directly attributable to or dependent upon the estuaries in these states. Based on commercial landings in Maryland and Virginia, the values for three key species—blue crab, striped bass, and menhaden- totaled nearly \$69 million in 2007 in the Chesapeake Bay alone.

Assessing how eutrophication in estuaries affects fishery resources requires bioeconomic models (i.e., models that combine biological models of fish population dynamics with economic models describing fish harvesting and consumption decisions), but relatively few exist (Knowler, 2002). Kahn and Kemp (1985) estimated that a 50% decline in submerged aquatic vegetation (SAV) from levels existing in the late 1970s (similar to current levels [Chesapeake Bay Program, 2008]) would decrease the net social benefits from striped bass by \$16 million (in 2007 dollars). In a separate analysis, Anderson (1989) modeled blue crab harvests under baseline conditions and under conditions with “full restoration” of SAV. In equilibrium, the increase in annual producer surplus and consumer surplus with full restoration of SAV was estimated to be \$7.9 million (in 2007 dollars) or an 11% increase from current service provision from blue crab alone. Mistiaen et al. (2003) found that reductions in dissolved oxygen (DO) cause a statistically significant reduction in commercial harvest and revenues crab harvests. For the Patuxent River alone, a simulated reduction of DO from 5.6 to 4.0 mg/L was estimated to reduce crab harvests by 49% and reduce total annual earnings in the fishery by \$275,000 (in 2007 dollars). While these values do not quantify the increase in terms of atmospheric loadings alone, the estimated 20% loading to the Potomac River watershed (**REA 5.2.4**) from atmospheric deposition indicates that the benefits apportioned to deposition are significant.

In addition, eutrophication in estuaries may also affect the demand for seafood. For example, a well-publicized toxic pfiesteria bloom in the Maryland Eastern Shore in 1997 led to an estimated \$56 million (in 2007 dollars) in lost seafood sales for 360 seafood firms in

1 Maryland in the months following the outbreak (Lipton, 1999). Surveys by Whitehead, Haab,  
2 and Parsons (2003) and Parsons et al. (2006) indicated a reduction in consumer surplus due to  
3 eutrophication-related fish kills ranging from \$2 to \$5 per seafood meal.<sup>8</sup> As a result, they  
4 estimated aggregate consumer surplus losses of \$43 million to \$84 million (in 2007 dollars) in  
5 the month after a fish kill.

6 As mentioned in the REA (**5.2.1.3**), estuaries in the eastern United States also provide an  
7 important and substantial variety of cultural ecosystem services, including water-based  
8 recreational and aesthetic services. For example, FHWAR data indicate that 4.8% of the  
9 population in coastal states from North Carolina to Massachusetts participated in saltwater  
10 fishing, with a total of 26 million saltwater fishing days in 2006 (U.S. DOI, 2007). Based on  
11 estimates in **Section 5.2.1.3 of the REA**, total recreational consumer surplus value from these  
12 saltwater fishing days was approximately \$1.3 billion (in 2007 dollars). Recreational  
13 participation estimates for several other coastal recreational activities are also available for  
14 1999–2000 from the NSRE. Almost 6 million individuals participated in motorboating in coastal  
15 states from North Carolina to Massachusetts. Again, based on analysis in the REA, the aggregate  
16 value of these coastal motorboating outings was \$2 billion per year. Almost 7 million people  
17 participated in birdwatching, for a total of almost 175 million days per year, and more than 3  
18 million participated in visits to nonbeach coastal waterside areas, for a total of more than 35  
19 million days per year.

20 Estuaries and marshes have the potential to support a wide range of regulating services,  
21 including climate, biological, and water regulation; pollution detoxification; erosion prevention;  
22 and protection against natural hazards (MEA, 2005c). The relative lack of empirical models and  
23 valuation studies imposes obstacles to the estimation of ecosystem services affected by nitrogen  
24 deposition. While atmospheric deposition contributes to eutrophication there is uncertainty in  
25 separating the effects of atmospheric nitrogen from nitrogen reaching the estuaries from many  
26 other sources.

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<sup>8</sup> Surprisingly, these estimates were not sensitive to whether the fish kill was described as major or minor or to the different types of information included in the survey.

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2  
3

**Table 3-7. Summary of Values for Current Levels of Services and Changes in Service Levels in \$2007**

<b>Ecosystem Service</b>	<b>Area or Population Affected</b>	<b>Value (\$2007)</b>	<b>Species</b>
Total Catch – Commercial Fishing	14 east coast states	\$1.5 b/yr	
	MD/VA	\$69 m/yr	blue crab, striped bass, menhaden
<b>Change in Ecosystem Service</b>		<b>Value of Change (\$2007)</b>	
50% decline in SAV	Chesapeake Bay	↓ \$16 m/yr	striped bass
Full restoration SAV	Chesapeake Bay	↑ \$ 8 m/yr	blue crab
0.4% mg/L decrease DO	Patuxent River	↓ \$275 th/yr	↓ 49% blue crab harvest
HAB	1997 MD eastern shore	↓ \$56 m	loss to seafood industry
		↓ \$43-84 m	sustained loss over months
<b>Ecosystem Service</b>		<b>Participation Days</b>	<b>Value (\$2007)</b>
Saltwater fishing	4.6% pop. MA-NC	26m days	\$1.3 b/yr
Motorboating	6 million		2 b/yr
Bird watching	7 million	175 m days	
Non-beach coastal visits	3 million	35 m days	

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1 **3.4.3.1 Value of aquatic ecosystem service impairments from current levels nutrient**  
 2 **enrichment**

3 The aquatic nutrient enrichment case study relied on the NOAA Eutrophication Index as  
 4 the indicator, which includes dissolved oxygen, HABs, loss of SAV and loss of water clarity.  
 5 There are methods available to link some of the components to ecosystem services, most notably  
 6 loss of SAV and reductions in DO. The REA analysis estimates the change in several ecosystem  
 7 services including recreational fishing, boating, beach use, aesthetic services and nonuse  
 8 services. The REA focuses on two major East Coast estuaries – the Chesapeake Bay and the  
 9 Neuse River. Both estuaries receive between 20%-30% percent of their annual nitrogen loadings  
 10 through atmospheric deposition and both are showing symptoms of eutrophication. The analysis  
 11 uses and adapts results from several existing studies to approximate effects on several ecosystem  
 12 services, including commercial fishing, recreation, aesthetic enjoyment, and nonuse values. For  
 13 example, it is estimated that atmospheric nitrogen decreases the annual benefits of recreational  
 14 fishing, boating, and beach use in the Chesapeake Bay by \$43-\$217 million, \$3-8 million, and  
 15 \$124 million respectively, and reduces annual aesthetic benefits to nearshore residents by \$39-  
 16 102 million. In the Neuse River, the value of annual commercial crab fishing services would be  
 17 between \$0.1-1 million higher without the contribution of atmospheric nitrogen, and recreation  
 18 fishing services in the larger Albermarle Pamlico Sound estuary system (which includes the  
 19 Neuse) would be \$1-8 million greater per year.

20

21 **Table 3-8. Summary of Annual Damages to Services due to Atmospheric Loading**

<b>Ecosystem Service</b>	<b>Annual Value (\$2007)</b>	<b>Waterbody Affected</b>
Recreational Saltwater Fishing	\$43-217 b	Chesapeake Bay
	\$1-8 m	Albemarle Pamlico Sound
Beach Use	\$39-102 m	Chesapeake Bay
Boating	\$3-8 m	Chesapeake Bay
Commercial Crab Fishing	\$0.1-1 m	Neuse River

22

23

24

#### 1 **3.4.4 Evidence for Adversity Related to Terrestrial Nutrient Enrichment**

2 The ecosystem service impacts of terrestrial nutrient enrichment in unmanaged ecosystems  
3 include primarily cultural and regulating services. In CSS areas, concerns focus on a decline in  
4 CSS and an increase in nonnative grasses and other species, impacts on the viability of threatened  
5 and endangered species associated with CSS, and an increase in fire frequency. Changes in MCF  
6 include changes in habitat suitability and increased tree mortality, increased fire intensity, and a  
7 change in the forest's nutrient cycling that may affect surface water quality through nitrate  
8 leaching (EPA, 2008).

9 The terrestrial nutrient enrichment case study relies on benchmark deposition levels for  
10 various species and ecosystems as indicators of ecosystem response. While it would be expected  
11 that deposition above those levels would have deleterious effects on the provision of ecosystem  
12 services in those areas, at this time it is possible only to describe the magnitude of the some of  
13 the services currently being provided. Methods are not yet available to allow estimation of  
14 changes in services due to nitrogen deposition.

15 The value that California residents and the U.S. population as a whole place on CSS and  
16 MCF habitats is reflected in the various federal, state, and local government measures that have  
17 been put in place to protect these habitats. Threatened and endangered species are protected by  
18 the Endangered Species Act. The State of California passed the Natural Communities  
19 Conservation Planning Program (NCCP) in 1991, and CSS was the first habitat identified for  
20 protection under the program (see [www.dfg.ca.gov/habcon/nccp](http://www.dfg.ca.gov/habcon/nccp)). It is estimated that only 10 –  
21 15% of the original extent of CSS habitat remains ([NPS.gov/cabr/naturescience/coastal-sage-scrub-and-southern-chaparral-communities.htm](http://NPS.gov/cabr/naturescience/coastal-sage-scrub-and-southern-chaparral-communities.htm)). Private organizations such as The Nature  
22 Conservancy, the Audubon Society, and local land trusts also protect and restore CSS and MCF  
23 habitat.  
24

25 CSS and MCF are found in numerous recreation areas in California. Three national parks  
26 and monuments in California contain CSS, including Cabrillo National Monument, Channel  
27 Islands National Park, and Santa Monica National Recreation Area. All three parks showcase  
28 CSS habitat with educational programs and information provided to visitors, guided hikes, and  
29 research projects focused on understanding and preserving CSS. Over a million visitors traveled  
30 through these three parks in 2008. MCF is highlighted in Sequoia and Kings Canyon National

1 Park, Yosemite National Park, and Lassen Volcanic National Park, where more than 5 million  
 2 people visited in 2008.

3 The 2006 FHWAR for California (DOI, 2007) reports on the number of individuals  
 4 involved in fishing, hunting, and wildlife viewing in California. Millions of people are involved  
 5 in just these three activities each year. The quality of these trips depends in part on the health of  
 6 the ecosystems and their ability to support the diversity of plants and animals found in important  
 7 habitats found in CSS or MCF ecosystems and the parks associated with those ecosystems.  
 8 Based on analyses in **Section 5.3.1.3 of the REA** (U.S.EPA, 2009), average values of the total  
 9 benefits in 2006 from fishing, hunting, and wildlife viewing away from home in California were  
 10 approximately \$947 million, \$169 million, and \$3.59 billion, respectively. In addition, data from  
 11 California State Parks (2003) indicate that in 2002, 68.7% of adult residents participated in trail  
 12 hiking for an average of 24.1 days per year. The analyses in the REA (U.S.EPA, 2009) indicate  
 13 that the aggregate annual benefit for California residents from trail hiking in 2007 was \$11.59  
 14 billion. It is not currently possible to quantify the loss in value of services due to nitrogen  
 15 deposition as those losses are already reflected in the estimates of the contemporaneous total value  
 16 of these recreational activities. Restoration of services through decreases in nitrogen deposition  
 17 would likely increase the total value of recreational services.

18  
 19 **Table 3-9. Summary of Current Levels of Ecosystem Services**

<b>Activity</b>	<b>Participation</b>	<b># of days/yr</b>	<b>Average WTP</b>	<b>Annual Aggregate Value (\$2007 in millions)</b>
<b>Trail Hiking</b>	68.7% of CA population	453 m	\$25.59	115,900
<b>Fishing</b>	1.7 m	19 m	\$48.86	947
<b>Wildlife Viewing</b>	6.2 m	45 m	\$79.81	3,600
<b>Hunting</b>	0.28 m	3.3 m	\$50.10	169

20 Sources: 2006 FHWAR for California (DOI, 2007), California State Parks (2003),  
 21 Kaval and Loomis(2003)  
 22

1 CSS and MCF are home to a number of important and rare species and habitat types. CSS  
2 displays richness in biodiversity with more than 550 herbaceous annual and perennial species. Of  
3 these herbs, nearly half are endangered, sensitive, or of special status (Burger et al., 2003).  
4 Additionally, avian, arthropod, herpetofauna, and mammalian species live in CSS habitat or use  
5 the habitat for breeding or foraging. Communities of CSS are home to three important federally  
6 endangered species: the Quino checkerspot butterfly, the kangaroo rat and the California  
7 gnatcatcher. MCF is home to one federally endangered species (mountain yellow-legged frog)  
8 and a number of state-level sensitive species. The Audubon Society lists 28 important bird areas  
9 in CSS habitat and at least 5 in MCF in California (<http://ca.audubon.org/iba/index.shtml>).<sup>9</sup>

10 The terrestrial enrichment case study in **Section 5.3.1.3** of the REA and **Section 3.3.5** of  
11 the ISA identified fire regulation as a service that could be affected by nutrient enrichment of the  
12 CSS and MCF ecosystems by encouraging growth of more flammable grasses, increasing fuel  
13 loads, and altering the fire cycle. Over the 5-year period from 2004 to 2008, Southern California  
14 experienced, on average, over 4,000 fires per year burning, on average, over 400,000 acres per  
15 year (National Association of State Foresters [NASF], 2009). It is not possible at this time to  
16 quantify the contribution of nitrogen deposition, among many other factors, to increased fire risk.

17 The CSS and MCF were selected as case studies for terrestrial enrichment because of the  
18 potential that these areas could be adversely affected by excessive N deposition. To date, the  
19 detailed studies needed to identify the magnitude of the adverse impacts due to N deposition  
20 have not been completed. Based on available data, this report provides a qualitative discussion of  
21 the services offered by CSS and MCF and a sense of the scale of benefits associated with these  
22 services. California is famous for its recreational opportunities and beautiful landscapes. CSS  
23 and MCF are an integral part of the California landscape, and together the ranges of these  
24 habitats include the densely populated and valuable coastline and the mountain areas. Through  
25 recreation and scenic value, these habitats affect the lives of millions of California residents and  
26 tourists. Numerous threatened and endangered species at both the state and federal levels reside  
27 in CSS and MCF. Both habitats may play an important role in wildfire frequency and intensity,  
28 an extremely important problem for California. The potentially high value of the ecosystem  
29 services provided by CSS and MCF justify careful attention to the long-term viability of these  
30 habitats.

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<sup>9</sup> Important Bird Areas are sites that provide essential habitat for one or more species of bird.

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## **4 ADDRESSING THE ADEQUACY OF THE CURRENT STANDARDS**

Based on the information in Chapters 2 and 3, we conclude that there is support in the available effects-based evidence for consideration of secondary standards for NO<sub>x</sub> and SO<sub>x</sub> that are protective against adverse ecological effects associated with deposition of NO<sub>x</sub> and SO<sub>x</sub> to sensitive ecosystems. Having reached this general conclusion, we then to the extent possible evaluate the adequacy of the current NO<sub>x</sub> and SO<sub>x</sub> secondary standards by considering to what degree risks to sensitive ecosystems would be expected to occur in areas that meet the current standards. Staff conclusions regarding the adequacy of the current standards are based on the available ecological effects, exposure and risk-based evidence. In evaluating the strength of this information, staff have taken into account the uncertainties and limitations in the scientific evidence. This chapter addresses key policy relevant questions that inform our determination regarding the adequacy of the structure and levels of the current secondary standards. The chapter begins with a discussion of the structure of the current standards, followed by a presentation of information on recent air quality relative to the existing standards, recent NO<sub>x</sub> and SO<sub>x</sub> deposition levels, evaluation of recent deposition levels relative to levels where adverse ecological effects have been observed, and a set of conclusions regarding the adequacy of the current structure and levels of the standards. Acidification occurs over extended periods and the ability of both terrestrial and aquatic systems to recover is dependent upon not only the decrease in acidic deposition, but the ability of these ecosystems to generate cations needed for nutrients and base cation supply. As a result, given the same decrease in deposition, ecosystems with high levels of base cation replacement will recover faster than those with low levels.

### **4.1 Are The Structures Of The Current NO<sub>x</sub> And SO<sub>x</sub> Secondary Standards Based On Relevant Ecological Indicators Such That They Are Adequate To Determine And Protect Public Welfare Against Adverse Effects On Ecosystems?**

The current secondary NO<sub>x</sub> and SO<sub>x</sub> standards are intended to protect against adverse effects to public welfare. For NO<sub>x</sub>, the current secondary standard was set identical to the

1 primary standard<sup>1</sup>, e.g. an annual standard set for NO<sub>2</sub> to protect against adverse effects on  
2 vegetation from direct exposure to ambient NO<sub>x</sub>. For SO<sub>x</sub>, the current secondary standard is a  
3 3-hour standard intended to provide protection for plants from the direct foliar damage  
4 associated with atmospheric concentrations of SO<sub>2</sub>. It is appropriate in this review to consider  
5 whether the current standards are adequate to protect against the direct effects on vegetation  
6 resulting from ambient NO<sub>2</sub> and SO<sub>2</sub> which were the basis for the current secondary standards.  
7 The ISA concluded that there was sufficient evidence to infer a causal relationship between  
8 exposure to SO<sub>2</sub>, NO, NO<sub>2</sub> and PAN and injury to vegetation. Additional research on acute  
9 foliar injury has been limited and there is no evidence to suggest foliar injury below the levels of  
10 the current secondary standards for SO<sub>x</sub> and NO<sub>x</sub>. There is sufficient evidence to suggest that  
11 the levels of the current standards are likely adequate to protect against direct phytotoxic effects.

12 The ISA however, has established that the major effects of concern for this review of the  
13 NO<sub>x</sub> and SO<sub>x</sub> standards are associated with deposition of N and S caused by atmospheric  
14 concentrations of NO<sub>x</sub> and SO<sub>x</sub> (see Chapter 2). As discussed in the following sections, the  
15 current standards are not directed toward depositional effects, and none of the elements of the  
16 current NAAQS – indicator, form, averaging time, and level – are suited for addressing the  
17 effects of N and S deposition. Thus, by using atmospheric NO<sub>2</sub> and SO<sub>2</sub> concentrations as  
18 indicators, the current standards address only a fraction of total atmospheric NO<sub>x</sub> and SO<sub>x</sub>, and  
19 do not take into account the effects from deposition of total atmospheric NO<sub>x</sub> and SO<sub>x</sub>. By  
20 addressing short-term concentrations, the current SO<sub>2</sub> standard, while protective against direct  
21 foliar effects from gaseous SO<sub>x</sub>, does not take into account the findings of effects in the ISA,  
22 which notes the relationship between annual deposition of S and acidification effects which are  
23 likely to be more severe and widespread than phytotoxic effects under current ambient  
24 conditions, and include effects from long term deposition as well as short term.. Acidification is  
25 a process which occurs over time, as the ability of an aquatic system to counteract acidic inputs  
26 is reduced as natural buffers are used more rapidly than they can be replaced through geologic  
27 weathering. The relevant period of exposure for ecosystems is therefore not the exposures  
28 captured in the short averaging time of the current SO<sub>2</sub> standard.

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<sup>1</sup> The current primary NO<sub>2</sub> standard has recently been changed to the 3 year average of the 98<sup>th</sup> percentile of the annual distribution of the 1 hour daily maximum of the concentration of NO<sub>2</sub>. The current secondary standard remains as it was set in 1971.

1           The levels of the current standards also are not well suited to dealing with deposition-  
2 based effects of NO<sub>x</sub> and SO<sub>x</sub>. Current standards are specified as allowable single atmospheric  
3 concentration levels for NO<sub>2</sub> or SO<sub>2</sub>. This type of structure does not take into account variability  
4 in the atmospheric and ecological factors that may alter the effects of NO<sub>x</sub> and SO<sub>x</sub> on public  
5 welfare. Consistent with section 108 of the CAA, the ISA includes in the air quality criteria  
6 consideration of how these variable factors impact the effects of ambient NO<sub>x</sub> and SO<sub>x</sub> on public  
7 welfare. See CAA section 108 (a)(2)(A) requiring air quality criteria to include information on  
8 “those variable factors (including atmospheric conditions) which of themselves or in  
9 combination with other factors may alter the effects on ... welfare of such air pollutant”.

10 Secondary standards are intended to address a wide variety of effects occurring in different types  
11 of environments and ecosystems. Ecosystems are not uniformly distributed either spatially or  
12 temporally in their sensitivity to air pollution. Therefore, failure to account for the major  
13 determinants of variability, including geological and soil characteristics related to the sensitivity  
14 to acidification as well as atmospheric and landscape characteristics that govern rates of  
15 deposition, may lead to standards that do not provide requisite levels of protection across  
16 ecosystems. Finally, given the mismatch of all of the other elements of the current secondary  
17 NAAQS with deposition-based effects, the form of those standards will also be mismatched.

18           Because most areas of the U.S. are in attainment with the current NO<sub>2</sub> and SO<sub>x</sub> standards,  
19 it is possible to evaluate current conditions, and evaluate the impact on public welfare from the  
20 current effects on ecosystems from NO<sub>x</sub> and SO<sub>x</sub> deposition in areas that attain the current  
21 standards that use NO<sub>2</sub> and SO<sub>2</sub> as indicators. In addition, this chapter qualitatively addresses  
22 the adequacy of the structures of the existing standards relative to ecologically relevant standards  
23 for NO<sub>x</sub> and SO<sub>x</sub>, and sets up arguments for developing an ecologically relevant structure for the  
24 standards as described in Chapter 5.

25

#### 26 **4.2 To What Extent Are The Structures Of The Current NO<sub>x</sub> AND SO<sub>x</sub> Secondary** 27 **Standards Meaningfully Related To Relevant Ecological Indicators Of Public** 28 **Welfare Effects?**

29

30           The current secondary standard for NO<sub>x</sub>, set in 1971, using NO<sub>2</sub> as the atmospheric  
31 indicator, is 0.053 parts per million (ppm) (100 micrograms per cubic meter of air [µg/m<sup>3</sup>]),

1 annual arithmetic average, calculated as the arithmetic mean of the 1-hour NO<sub>2</sub> concentrations.  
2 This standard was selected to provide protection to the public welfare against acute injury to  
3 vegetation from direct exposure and resulting phytotoxicity. During the last review of the NO<sub>x</sub>  
4 standards, impacts associated with chronic acidification and eutrophication from NO<sub>x</sub> deposition  
5 were acknowledged, but the relationships between atmospheric concentrations of NO<sub>x</sub> and levels  
6 of acidification and eutrophication and associated welfare impacts were determined to be too  
7 uncertain to be useful as a basis for setting a national secondary standard (USEPA 1995).

8 The current secondary standard for SO<sub>x</sub>, set in 1971, uses SO<sub>2</sub> as the atmospheric  
9 indicator, is a 3-hour average of 0.5 ppm, not to be exceeded more than once per year. This  
10 standard was selected to provide protection to the public welfare against acute injury to  
11 vegetation. In the last review of the SO<sub>x</sub> secondary standard, impacts associated with chronic  
12 acidification were acknowledged, but the relationships between atmospheric concentrations of  
13 SO<sub>x</sub> and levels of acidification, along with the complex interactions between SO<sub>x</sub> and NO<sub>x</sub> in  
14 acidification processes, were cited as critical uncertainties which made the setting of secondary  
15 NAAQS to protect against acidification inappropriate at that time (USEPA 1982).

16 In the previous separate reviews of the NO<sub>x</sub> and SO<sub>x</sub> secondary standards, EPA  
17 acknowledged in each review the additional impacts of NO<sub>x</sub> and SO<sub>x</sub> on public welfare through  
18 the longer term impact of the pollutants once deposited to ecosystems. However, the previous  
19 reviews cited numerous uncertainties as the basis for not directly addressing those impacts in the  
20 setting of secondary standards. In addition, these previous reviews did not consider the common  
21 pathways of impact for both nitrogen and sulfur acting on the same ecosystem endpoints.

22 Three issues arise that call into question the ecological relevance of the current structure  
23 of the secondary standards for NO<sub>x</sub> and SO<sub>x</sub>. One issue is the exposure period that is relevant  
24 for ecosystem impacts. The majority of deposition related impacts are associated with  
25 depositional loads that occur over periods of months to years. This differs significantly from  
26 exposures associated with hourly concentrations of NO<sub>2</sub> and SO<sub>2</sub> as measured by the current  
27 standards. Even though the NO<sub>2</sub> standard uses an annual average of NO<sub>2</sub>, it is focused on the  
28 annual average of 1-hour NO<sub>2</sub> concentrations, rather than on a cumulative metric or an averaging  
29 metric based on daily or monthly averages. A second issue is the choice of atmospheric  
30 indicators. NO<sub>2</sub> and SO<sub>2</sub> are used as the component of oxides of nitrogen and sulfur that are  
31 measured, but they do not provide a complete link to the direct effects on ecosystems from

1 deposition of NO<sub>x</sub> and SO<sub>x</sub> as they do not capture all relevant chemical species of oxidized  
2 nitrogen and oxidized sulfur that contribute to deposition. The ISA provides evidence that  
3 deposition related effects are linked with total nitrogen and total sulfur deposition, and thus all  
4 forms of oxidized nitrogen and oxidized sulfur that are deposited will contribute to effects on  
5 ecosystems. This suggests that more comprehensive atmospheric indicators should be  
6 considered in designing ecologically relevant standards. Further discussions of the need for  
7 more ecologically relevant atmospheric indicators as well as the relative contributions to  
8 deposition from various species of NO<sub>x</sub> and SO<sub>x</sub> can be in found in Chapters 5 and 6 below.  
9 The third issue is that the current standards reflect separate assessments of the two individual  
10 pollutants, NO<sub>2</sub> and SO<sub>2</sub>, rather than assessing the joint impacts of deposition of NO<sub>x</sub> and SO<sub>x</sub> to  
11 ecosystems, recognizing the role that each pollutant plays in jointly affecting ecosystem  
12 indicators, functions, and services. The clearest example of this interaction is in assessment of  
13 the impacts of acidifying deposition on aquatic ecosystems.

14         Acidification in an aquatic ecosystem depends on the total acidifying potential of the  
15 deposition of both N and S from both atmospheric deposition of NO<sub>x</sub> and SO<sub>x</sub> as well as the  
16 inputs from other sources of N and S such as reduced nitrogen and non-atmospheric sources. It is  
17 the joint impact of the two pollutants that determines the ultimate effect on organisms within the  
18 ecosystem, and critical ecosystem functions such as habitat provision and biodiversity.  
19 Standards that are set independently are less able to account for the contribution of the other  
20 pollutant. This suggests that interactions between NO<sub>x</sub> and SO<sub>x</sub> should be a critical element of  
21 the conceptual framework for ecologically relevant standards. There are also important  
22 interactions between NO<sub>x</sub> and SO<sub>x</sub> and reduced forms of nitrogen, which also contribute to  
23 acidification and nutrient enrichment. Although the standards do not directly address reduced  
24 forms of nitrogen in the atmosphere, e.g. they do not require specific levels of reduced nitrogen,  
25 it is important that the structure of the standards address the role of reduced nitrogen in  
26 determining the ecological effects resulting from deposition of atmospheric NO<sub>x</sub> and SO<sub>x</sub>.  
27 Consideration will also have to be given to account for loadings coming from non-atmospheric  
28 sources as ecosystems will respond to these sources as well.

29         In addition to the fundamental issues discussed above, the current structures of the  
30 standards do not address the complexities in the responses of ecosystems to deposition of NO<sub>x</sub>  
31 and SO<sub>x</sub>. Ecosystems contain complex groupings of organisms that respond in various ways to

1 the alterations of soil and water that result from deposition of nitrogen and sulfur compounds.  
2 Different ecosystems therefore respond in different ways depending on a multitude of factors  
3 that control how deposition is integrated into the system. For example, the same levels of  
4 deposition falling on limestone dominated soils have a very different effect than those falling on  
5 shallow glaciated soils underlain with granite. One system may over time display no obvious  
6 detriment while the other may experience a catastrophic loss in fish communities. This degree  
7 of sensitivity is a function of many atmospheric factors which control rates of deposition as well  
8 as ecological factors which control how an ecosystem responds to that deposition. The current  
9 standards do not take into account spatial and seasonal variations not only in depositional  
10 loadings but also in sensitivity of ecosystems exposed to those loadings.

11  
12 **4.3 To What Extent Do Current Monitoring Networks Provide A Sufficient Basis For**  
13 **Determining The Adequacy of Current Secondary NO<sub>x</sub> and SO<sub>x</sub> Standards?**

14  
15 Staff have closely evaluated whether levels of N and S allowed by current standards are  
16 requisite to protect public welfare. Doing so requires relating atmospheric concentrations with  
17 deposition, exposure pathways, and measured effects on ecologic receptors. A combination of  
18 monitoring and air quality model applications is useful in linking atmospheric concentrations to  
19 ecological effects. There are over 1000 ground level monitoring platforms (Figures 4-1 and 4-2  
20 and Table 4 -1) that provide measurements of some form of atmospheric nitrogen or sulfur. The  
21 key pollutants for this assessment are total oxidized nitrogen (NO<sub>y</sub>), total reduced nitrogen  
22 (NH<sub>x</sub>), and total oxidized sulfur which is referenced herein as (SO<sub>x</sub>) and defined as the sum of  
23 SO<sub>2</sub> (gas) and particulate sulfate.. Total reactive oxidized atmospheric nitrogen, NO<sub>y</sub>, is defined  
24 as NO<sub>x</sub> (NO and NO<sub>2</sub>) and all oxidized NO<sub>x</sub> products: NO<sub>y</sub> = NO<sub>2</sub> + NO + HNO<sub>3</sub> + PAN  
25 +2N<sub>2</sub>O<sub>5</sub> + HONO+ NO<sub>3</sub> + organic nitrates + particulate NO<sub>3</sub> (Finlayson-Pitts and Pitts, 2000).  
26 This definition of NO<sub>y</sub> reflects the operational principles of standard measurement techniques in  
27 which all oxidized nitrogen species are converted to nitrogen oxide (NO) through catalytic  
28 reduction and the resulting NO is detected through luminescence. Thus, NO<sub>y</sub> is truly defined as  
29 total oxidized nitrogen as converted to NO, essentially representing all oxidized nitrogen atoms.  
30 NO<sub>y</sub> is not a strict representation of the all moles of oxidized nitrogen as the diatomic  
31

**Table 4-1. Summary of Monitoring Networks.**

<b>Network</b>	<b>Number of Sites</b>	<b>Species Measured</b>	<b>Sampling Frequency</b>	<b>Comments</b>
<b>All Sulfur Sites</b>				
NCore	82	SO <sub>2</sub>	Hourly	Includes 20 rural sites
SEARCH	8	SO <sub>2</sub>	Hourly	Includes 3 rural sites
SO <sub>2</sub>	751	SO <sub>2</sub>	Hourly	NAMS/SLAMS/PAMS for 2008
PM Speciation	242	Sulfates	24-hour	Measurements of Sulfates (88403) identified in AQS for Trends and Supplemental Speciation monitoring type for 2008
IMPROVE	215	Sulfates	24-hour	IMPROVE Monitoring Sites with Measurements of Sulfates (88403) identified in AQS
CASTNET	88	Sulfates	Weekly Ave.	EPA & NPS

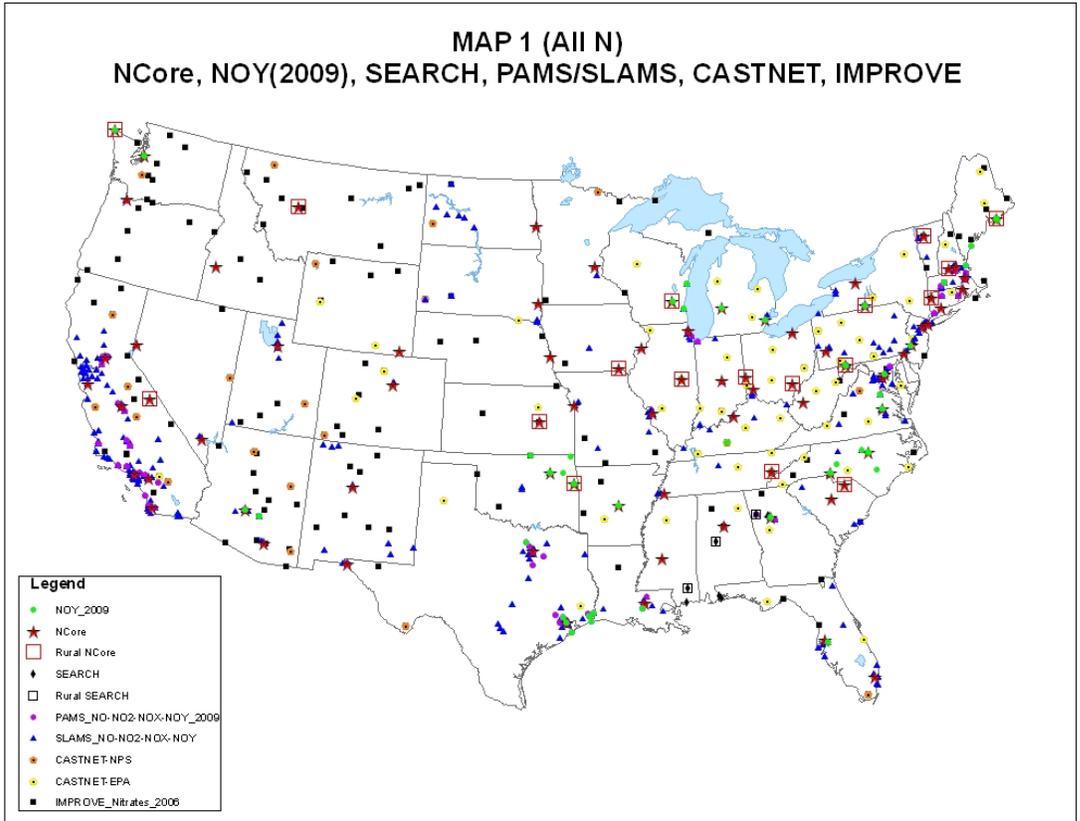
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2

**Table 4-1 Summary of Monitoring Networks (continued)**

<b>Network</b>	<b>Number of Sites</b>	<b>Species Measured</b>	<b>Sampling Frequency</b>	<b>Comments</b>
<b>All Nitrogen Sites</b>				
NCore	82	NO/NO <sub>y</sub>	Hourly	Includes 20 rural sites
SEARCH	8	NO/NO <sub>2</sub> /NO <sub>y</sub> /HNO <sub>3</sub>	Hourly	Includes 3 rural sites
PAMS	119	NO <sub>2</sub> /NO <sub>x</sub>	Hourly	Official sites as of 12/09
SLAMS	643	NO/NO <sub>2</sub> /NO <sub>x</sub> /NO <sub>y</sub>	Hourly	All SLAMS Monitoring Sites with Measurements of NO, NO <sub>2</sub> , NO <sub>x</sub> or NO <sub>y</sub> in 2009 identified in AQS
NOY	59	NO <sub>y</sub>	Hourly	All Monitoring Sites with Measurements of NO <sub>y</sub> in 2009 identified in AQS, regardless of Monitoring Type
IMPROVE	214	Nitrates	24-hour	IMPROVE Monitoring Sites with Measurements of Nitrates (88306) identified in AQS
CASTNET	88	Nitrates	Weekly Ave.	EPA & NPS

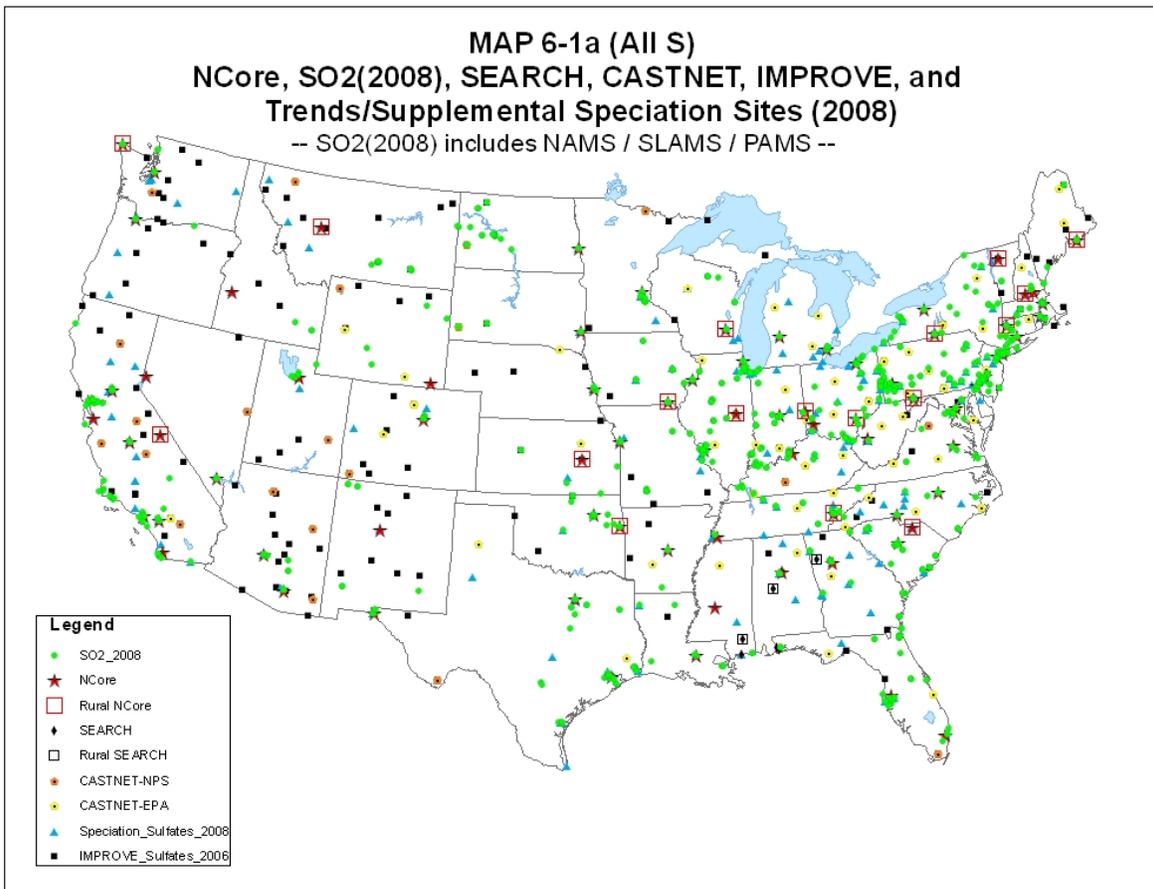
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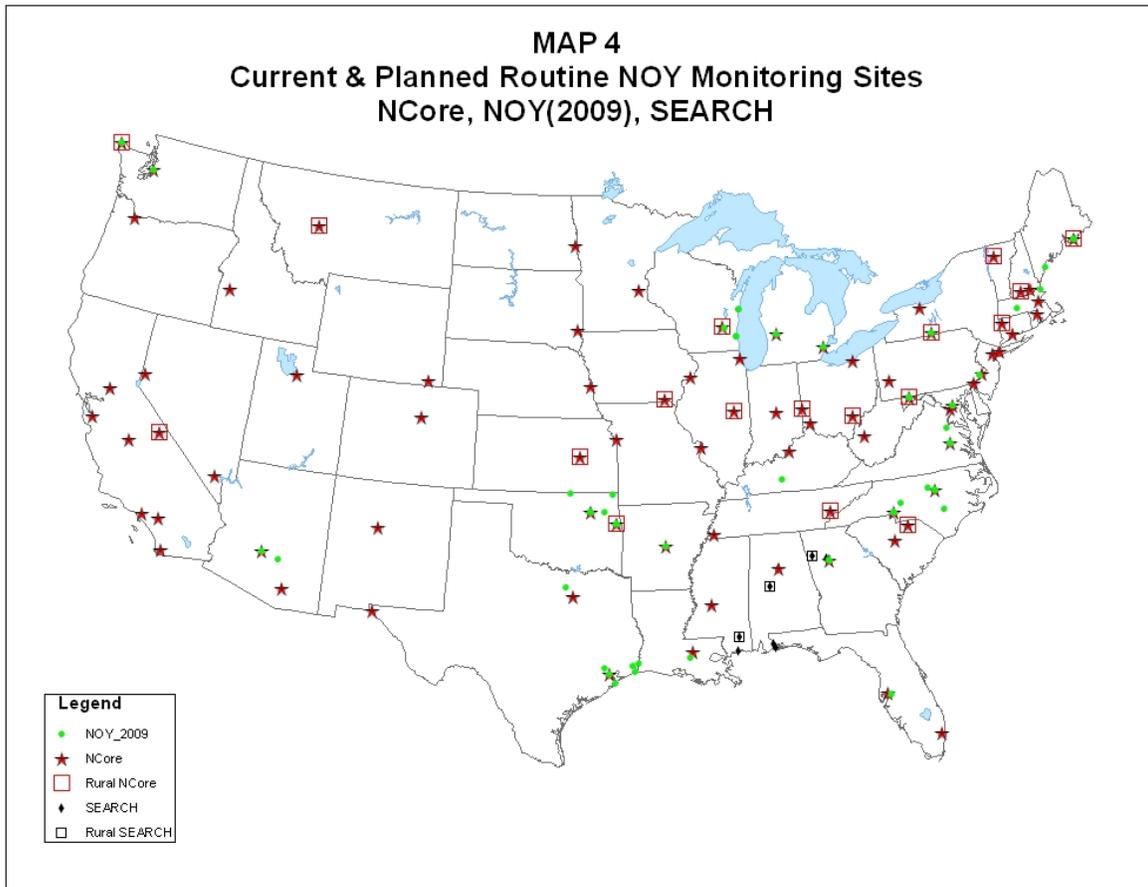
**Figure 4-1** Routinely operating surface monitoring stations measuring forms of atmospheric nitrogen.



1  
 2 **Figure 4-2** Routinely operating surface monitoring stations measuring forms of  
 3 **atmospheric sulfur.** All site locations measure both SO<sub>2</sub> and sulfate except for  
 4 the green SO<sub>2</sub> only sites.  
 5

1 nitrogen species such as  $N_2O_5$  yield 2 moles of NO. This definition is consistent with the  
2 relationship between atmospheric nitrogen and acidification processes as the reported  $NO_Y$   
3 provides a direct estimate of the potential equivalents available for acidification. Total reduced  
4 nitrogen ( $NH_X$ ) includes ammonia,  $NH_3$ , plus ammonium,  $NH_4$  (EPA, 2008). Reduced nitrogen  
5 plus oxidized nitrogen is referred to as total reactive nitrogen. Total oxidized sulfur ( $SO_X$ )  
6 includes  $SO_2$  gas and particulate sulfate,  $SO_4$ . These species are converted to mass of sulfur  
7 which is used directly, or converted to charge equivalents, in deposition analyses linking  
8 atmospheric deposition and ecosystem models. Ammonium and sulfate are components of  
9 atmospheric particulate matter as well as directly measured and modeled in precipitation as  
10 direct deposition components. As discussed in this section, there are only very limited routine  
11 measurements of total oxidized and reduced nitrogen. In addition, existing monitoring networks  
12 do not provide adequate geographic coverage to fully assess concentrations and deposition of  
13 reactive nitrogen and sulfur in and near sensitive ecosystems.

14 The principal monitoring networks include the regulatory based State and Local Air  
15 Monitoring Stations (SLAMS) providing mostly urban-based  $SO_2$ , NO and  $NO_X$ , the  $PM_{2.5}$   
16 chemical speciation networks Interagency Monitoring of Protected Visual Environments  
17 (IMPROVE) and EPA's Chemical Speciation Network (CSN) providing particle bound sulfate  
18 and nitrate, and the Clean Air Status and Trends Network (CASTNET) providing weekly  
19 averaged values of  $SO_2$ , nitric acid, and particle bound sulfate, nitrate and ammonium. The  
20 private sector supported South Eastern Aerosol Research and Characterization (SEARCH) Study  
21 network of 4-8 sites in the Southeast provides the only routinely operating source of true  
22 continuous  $NO_2$ , ammonia, and nitric acid measurements. SEARCH also provides  $PM_{2.5}$  size  
23 fractions of nitrate and sulfate. Collectively, the SLAMS, Photochemical Assessment  
24 Measurement Stations (PAMS), SEARCH and NCore networks will provide over 100 sites  
25 measuring  $NO_Y$  (Figure 4-3). The NCore network (Scheffe et al., 2009) is a multiple pollutant  
26 network with co-located measurements of key trace gases (CO,  $SO_2$ ,  $O_3$ , NO and  $NO_Y$ ),  $PM_{2.5}$   
27 and  $PM_{(10-2.5)}$  mass and  $PM_{2.5}$  chemical speciation. Additional air pollutants, particularly volatile  
28 organic compounds (VOCs), will be measured at those sites that are part of the existing PAMS  
29 and National Air Toxics Trends (NATTS) platforms. The NATTS (EPA, 2008) include 27  
30 stations across the U.S. that monitor for a variety of hazardous air pollutants and are intended to  
31 remain in place to provide a long-term record. Additional



1  
 2 **Figure 4-3. Anticipated network of surface based NO<sub>y</sub> stations based on 2009 network**  
 3 **design plans.** The NCore stations are scheduled to be operating by January,  
 4 2011.  
 5

6 measurements of ammonia and possibly true NO<sub>2</sub> are under consideration. True NO<sub>2</sub> is noted to  
 7 differentiate from the NO<sub>2</sub> determined through routine regulatory networks that have known  
 8 variable positive bias for NO<sub>2</sub>. The network currently is being deployed and expected to be  
 9 operational with nearly 75 sites by January, 2011. The sites are intended to serve as central site  
 10 monitors capturing broadly representative (e.g., not strongly influenced by nearby sources) air  
 11 quality in a suite of major and mid size cities, and approximately 20 sites are located in rural  
 12 locations.

13 There are significant measurement gaps for characterizing NO<sub>y</sub>, NH<sub>x</sub> and SO<sub>2</sub> in the  
 14 nations ambient air observation networks (EPA, 2008) that lead to greater reliance on air quality  
 15 modeling simulations to describe current conditions. National design of routinely operating  
 16 ambient air monitoring networks is driven mostly by data uses associated with implementing

1 primary NAAQS, with noted exceptions of the CASTNET and IMPROVE networks. In  
2 addition to significant spatial gaps in sensitive ecosystem areas that arise from a population  
3 oriented network design, the current measurements for primary and secondary nitrogen are  
4 markedly different and in some instances of negligible value for secondary NO<sub>x</sub> and SO<sub>x</sub>  
5 standards. For example, a true NO<sub>x</sub> (NO plus NO<sub>2</sub>) measurement typically would capture less  
6 than 50% (see discussion below) of the total regional NO<sub>y</sub> mass in rural locations as the more  
7 aged air masses contain significant oxidized nitrogen products in addition to NO<sub>x</sub>. Note that the  
8 NO<sub>x</sub> monitors used for NAAQS primary compliance purposes capture varying amounts of  
9 transformed nitrogen species; however, the method provides biased low estimates with  
10 significant airshed induced variability relative to true NO<sub>y</sub>. With the exception of the SEARCH  
11 network in the Southeast, there are virtually no routine networks that measure ammonia,  
12 although EPA is considering options for ammonia sampling in CASTNET and NCORE  
13 networks. Ammonium is reported in EPA chemical speciation networks, although the values are  
14 believed to be biased low due to ammonia volatilization.

15 CASTNET provides mostly rural measurements of SO<sub>2</sub>, total nitrate, and ammonium, and  
16 affords an existing infrastructure useful for future monitoring in support of a potential NO<sub>x</sub> and  
17 SO<sub>x</sub> secondary standard. However, the lack of NO<sub>y</sub>, SO<sub>x</sub> and NH<sub>x</sub> measurements in sensitive  
18 ecosystems will require attention in the N/S secondary standard proposal.

19 As a result of the limited monitoring networks for NO<sub>y</sub> and SO<sub>x</sub> in sensitive ecosystems,  
20 we are unable to use current ambient monitoring data to adequately link measured current  
21 atmospheric concentrations to ecological effects transmitted through deposition. At this time for  
22 the purpose of illustrating current atmospheric conditions, we supplement the available  
23 monitoring data with the use of sophisticated atmospheric modeling conducted using EPA's  
24 CMAQ model (as discussed in Chapter 7).

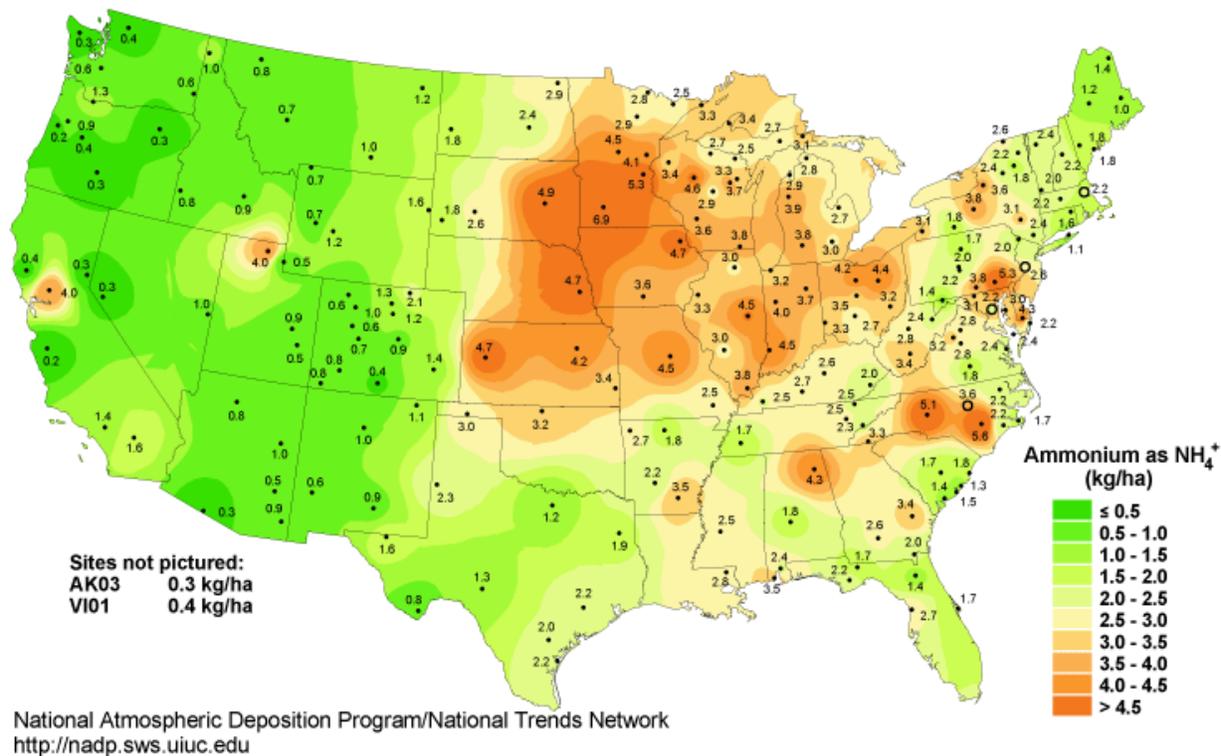
25

#### 26 **4.3.1 To what extent does the NADP monitoring network provide an adequate** 27 **characterization of deposition and what are the major limitations?**

28 The National Atmospheric Deposition Program (NADP) includes approximately 250 sites  
29 (Figure 4-4) across the U.S. providing annual total wet deposition based on weekly averaged  
30 measures of wet deposition of nitrate, ammonium, sulfate and other ions based on the  
31 concentrations of these ions in precipitation samples. Meteorological models have difficulty in

1 capturing the correct spatial and temporal features of precipitation events, raising the importance  
 2 of the NADP as a principal source of precipitation chemistry. The NADP has enabled several  
 3 organizations to participate in a measurement program with a centralized laboratory affording  
 4 measurement and analysis protocol consistency nationwide. Virtually every CASTNET site is  
 5 located at an NADP site and the combined NADP/CASTNET infrastructure is a starting point for  
 6 discussions addressing future NO<sub>x</sub> and SO<sub>x</sub> monitoring needs. The organic bound nitrogen is  
 7 not analyzed routinely in NADP samples. Consideration might be given to adding NADP sites  
 8 in locations where ambient air monitoring is conducted to assess compliance with a secondary  
 9 NO<sub>x</sub>/ SO<sub>x</sub> standard.  
 10

### Ammonium ion wet deposition, 2005



11  
 12 **Figure 4-4. Location of approximately 250 National Atmospheric Deposition Monitoring**  
 13 **(NADP) National Trends Network (NTN) sites illustrating annual**  
 14 **ammonium deposition for 2005.** Weekly values of precipitation based nitrate,  
 15 sulfate and ammonium are provided by NADP.  
 16  
 17

1 **4.3.2 What are the relative strengths and important gaps in existing atmospheric**  
2 **monitoring networks to address a combined NO<sub>x</sub>/SO<sub>x</sub> secondary standard?**

3  
4 Of the currently operating monitoring networks, precipitation based sulfate, ammonium  
5 and nitrate measurements provided by the NADP are the most relevant measurements that would  
6 support the secondary standard as they provide atmospheric deposition inputs that drive  
7 ecosystem models, and NADP site locations generally include acid sensitive areas. However,  
8 there are significant gaps in ambient air (aerosols and gases) monitoring networks for the  
9 measurement of the likely ambient indicators of NO<sub>y</sub>, SO<sub>2</sub>, and SO<sub>4</sub>. CASTNET filter packs  
10 provide the most relevant source of ambient sulfate (SO<sub>4</sub>) measurements as the open inlet of the  
11 filter packs incorporates the full range of particle sizes that contribute to deposition. The SO<sub>2</sub>  
12 measurements from CASTNET represent about 10% of all SO<sub>2</sub> sites nationally, but are  
13 especially relevant based on their locations in rural and regional settings, although CASTNET is  
14 not as spatially extensive (breadth and resolution) as the NADP network of precipitation sites.  
15 Although CASTNET does provide measurements of total ambient nitrate, other oxidized  
16 nitrogen species constituting a more complete NO<sub>y</sub> budget are not captured. In their current  
17 configuration, the State and local monitoring networks virtually offer no support for a secondary  
18 NO<sub>x</sub>/ SO<sub>x</sub> standard due to their urban based site orientation and exclusion of important oxidized  
19 nitrogen species (e.g., nitrates and PAN). The chemical speciation networks, including rural  
20 based IMPROVE, all provide ambient sulfate measurements based on on a 2.5μ size cut. While  
21 the sulfate mass within that size fraction may constitute 80% or greater of the ambient sulfate  
22 budget, the missing larger size particles can contribute significantly to sulfate deposition due to  
23 their relatively high gravitationally driven deposition velocities. Finally, there are virtually no  
24 ambient ammonia measurements routinely collected in acid sensitive areas. CASTNET does  
25 provide ammonium measurements, but the routine speciation networks that report ammonium  
26 have expected artifacts due to ammonia offgassing from nylon filters.

27  
28 Although this summary of existing networks suggests significant challenges in meeting  
29 the monitoring needs of a new NO<sub>x</sub>/ SO<sub>x</sub> standard, the networks do serve as a useful building  
30 block for moving forward. The site locations of NADP and CASTNET offer an infrastructure  
31 to accommodate additional instruments. The NCORE network has introduced nearly 75 NO<sub>y</sub>

1 trace level SO<sub>2</sub> monitors that are establishing operational familiarity and a basis for instrument  
2 performance characterization. In many cases, acid sensitive areas will be strongly influenced  
3 by regional transport of pollutants which typically is associated with relatively homogeneous  
4 spatial concentration patterns which allows for a correspondingly greater range of spatial  
5 representativeness of monitoring sites. Consequently, the expected burden on monitoring  
6 resources may be realistically dampened by the available infrastructure and expected  
7 homogeneity of air concentration patterns. A more thorough assessment of the adequacy of  
8 existing networks is predicated on identification of the area wide boundaries of the acid sensitive  
9 areas of concern which will initially be developed in the second PAD.

### 11 **4.3.3 How do we characterize deposition through monitoring and models?**

13 Routinely available directly measured precipitation to quantify wet deposition of sulfur  
14 and nitrogen species is provided through the NADP. Dry deposition is not a directly measured  
15 variable in routine monitoring efforts. It is important to pursue the development of direct dry  
16 deposition measurements to improve model parameterizations of deposition processes and  
17 possibly evolve into routine operations. Estimates of dry deposition based on observations are  
18 provided through the CASTNET program. However, dry deposition is a calculated value  
19 represented as the product of ambient concentration (either observed or estimated through air  
20 quality modeling) and deposition velocity,  $Dep_i^{Dry} = v_i^{Dry} \cdot C_i^{Amb}$

21 Deposition velocity is modeled as a mass transfer process through resistance layers  
22 associated with the canopy, uptake by vegetation, water and soil which collectively are  
23 influenced by micrometeorology, land surface and vegetation types and species specific  
24 solubility and reactivity. Dry deposition is calculated through deposition velocity models  
25 capturing these features and using species specific ambient air concentrations. This approach  
26 conceptually is similar using either observed or modeled air concentrations. Dry deposition  
27 estimates from the Community Multi-scale Air Quality (CMAQ) model (EPA, 1999) have been  
28 used in this assessment to provide spatially more resolved and extensive estimates of dry  
29 deposition for sulfur and all reactive nitrogen (oxidized and reduced) species (CASTNET does  
30 not capture important gases such as nitrogen dioxide, ammonia and peroxyacetyl nitrate). All  
31 of the relevant meteorological, land use, vegetation and elevation data required to estimate

1 deposition velocities are generated or accessible in the CMAQ and/or meteorological pre-  
2 processors.

3

4 **4.3.2.1 Why are we using CMAQ to model deposition? How are we using it? Why is**  
5 **CMAQ the right model to use? What is the spatial and temporal resolution of**  
6 **CMAQ? What are the model years? What are the limitations to CMAQ?**

7

8 CMAQ provides a platform that allows for a consistent mass accounting approach across  
9 ambient concentrations and dry and wet deposition values. Recognizing the limitations of  
10 ambient air networks, CMAQ was used to estimate dry deposition to complement NADP wet  
11 deposition for MAGIC modeling and for the first-order acidity balance (FAB) critical load  
12 modeling. CMAQ promotes analytical consistency and efficiency across analyses of multiple  
13 pollutants. EPA's Office of Research and Development continues to enhance the underlying  
14 deposition science in CMAQ. For the purposes of this policy assessment, CMAQ provides a  
15 consistent platform incorporating the atmospheric and deposition species of interest over the  
16 entire United States. The caveats and limitations of the use of model predictions are largely  
17 associated with the general reliance on calculated values, rather than on measurements. Model  
18 evaluation addressing the comparison of predictions with observed values is addressed in the  
19 REA and summarized in Chapter 7 of this PA. Currently, there are efforts to improve a number  
20 of nitrogen related processes in CMAQ, recognizing comparatively less uncertainty with the  
21 treatment of sulfur. Active areas of model process improvement are in the treatment of  
22 lightning generated NO<sub>x</sub> and the transference of nitrogen between atmospheric and terrestrial  
23 and aquatic media, often referred to as bi-directional flux. Lightning NO<sub>x</sub> potentially provides  
24 a significant contribution to wet deposition as the resulting NO<sub>x</sub> is rapidly entrained into aqueous  
25 cloud processes. Both the thermodynamics of soil processes and mass transfer of nitrogen  
26 species across the surface-atmosphere interface is governed by an assortment of temperature,  
27 moisture, advection and concentration patterns. These processes and mass transfer relationships  
28 are coupled within the emissions, meteorological, and chemical simulation processes and  
29 associated surface/vegetation and terrain information incorporated in or accessed by the CMAQ.  
30 In addition to research activities to improve the characterization of nitrogen-related processes in  
31 CMAQ, efforts are also underway to improve the general characterization of ammonia emissions

1 which remains as an area of large uncertainty due to limited source data and the ubiquitous  
2 nature of these emissions. Another challenge for regional/national air quality modeling is  
3 properly representing the effects on pollutant concentrations, precipitation and therefore  
4 deposition of variable terrain features, particularly steep mountain-valley gradients and the  
5 interfaces to wide open basins encountered in the Western United States.

6 The CMAQ was used in this assessment because it is the state of science model for  
7 simulating sources, formation, and fate of nitrogen and sulfur species. In addition to undergoing  
8 periodic independent scientific peer review, CMAQ bridges the scientific and regulatory  
9 communities as it is used extensively by EPA for regulatory air quality assessments and rules.  
10 CMAQ provides hourly estimates of the important precursor, intermediate and secondarily  
11 formed species associated with atmospheric chemistry and deposition processes influencing  
12 ozone, particulate matter concentrations and sulfur and nitrogen deposition. Simulations based  
13 on horizontal spatial scale resolutions of 12 km and 36 km were used in this policy assessment  
14 for 2002 – 2005.

#### 16 **4.4 What Is Our Best Characterization of Atmospheric Concentrations Of NO<sub>y</sub> and** 17 **SO<sub>x</sub>, and Deposition Of N And S?**

18  
19 Air quality models and blending of model results and observations are used to  
20 characterize current environmental state conditions due to the relative sparseness of monitoring  
21 coverage in sensitive ecosystems as well as gaps in coverage for specific atmospheric species of  
22 N and S most relevant to deposition, such as NO<sub>y</sub>, in available monitoring platforms.

##### 24 **4.4.1 What are the current atmospheric concentrations of reactive nitrogen, NO<sub>y</sub>,** 25 **reduced nitrogen, NH<sub>x</sub>, sulfur dioxide, SO<sub>2</sub>, and sulfate, SO<sub>4</sub>?**

26  
27 To provide information for use in characterizing the adequacy of the current standards, we assess  
28 the best available data for estimating the ambient concentrations of the major sources of  
29 atmospheric nitrogen and sulfur across the U.S. Acidification and nutrient enrichment processes  
30 are largely dependent on the cycling of total nitrogen and sulfur species. From an atmospheric  
31 perspective, it is convenient and consistent with current measurement and modeling frameworks

1 to consider the reduced and oxidized forms of atmospheric nitrogen. Virtually all atmospheric  
2 sulfur is considered oxidized sulfur in the forms of particulate bound sulfate and gaseous sulfur  
3 dioxide. In order to assess current concentrations of reactive nitrogen and sulfur, we evaluated  
4 data available from the existing monitoring networks as well as from the CMAQ model.  
5 Regarding the monitoring data, there are a number of important issues in understanding the  
6 measurements of  $\text{NO}_Y$  provided by different monitoring networks. In principle, measured  $\text{NO}_Y$  is  
7 based on catalytic conversion of all oxidized species to NO followed by chemiluminescence NO  
8 detection. We recognize the caveats associated with instrument conversion efficiency and  
9 possible inlet losses. The CMAQ treats the dominant  $\text{NO}_Y$  species as explicit species while the  
10 minor contributing non-PAN organic nitrogen compounds are aggregated. Atmospheric  
11 nitrogen and sulfur are largely viewed as regional air quality issues due to the importance of  
12 chemical conversion of primary emissions into secondarily formed species, a combination of  
13 ubiquitous sources, particularly mobile source emissions of  $\text{NO}_X$ , and elevated emissions of  $\text{NO}_X$   
14 and  $\text{SO}_2$  that aid pollutant mass dispersal and broader physical transport over large distances. In  
15 effect, the regional nature is due to both transport processes as well as the relatively ubiquitous  
16 nature of sources combined with chemical processes that tend to form more stable species with  
17 extended atmospheric lifetimes. This regionalized effect, particularly throughout the eastern  
18 United States, dominates the overall patterns discussed below of secondarily formed species such  
19 as sulfate or  $\text{NO}_Y$ , which is an aggregate of species with the more aged air masses consisting  
20 largely of chemically processed air dominated by secondarily formed peroxyacetyl nitrate  
21 (PAN), particulate nitrate and nitric acid.

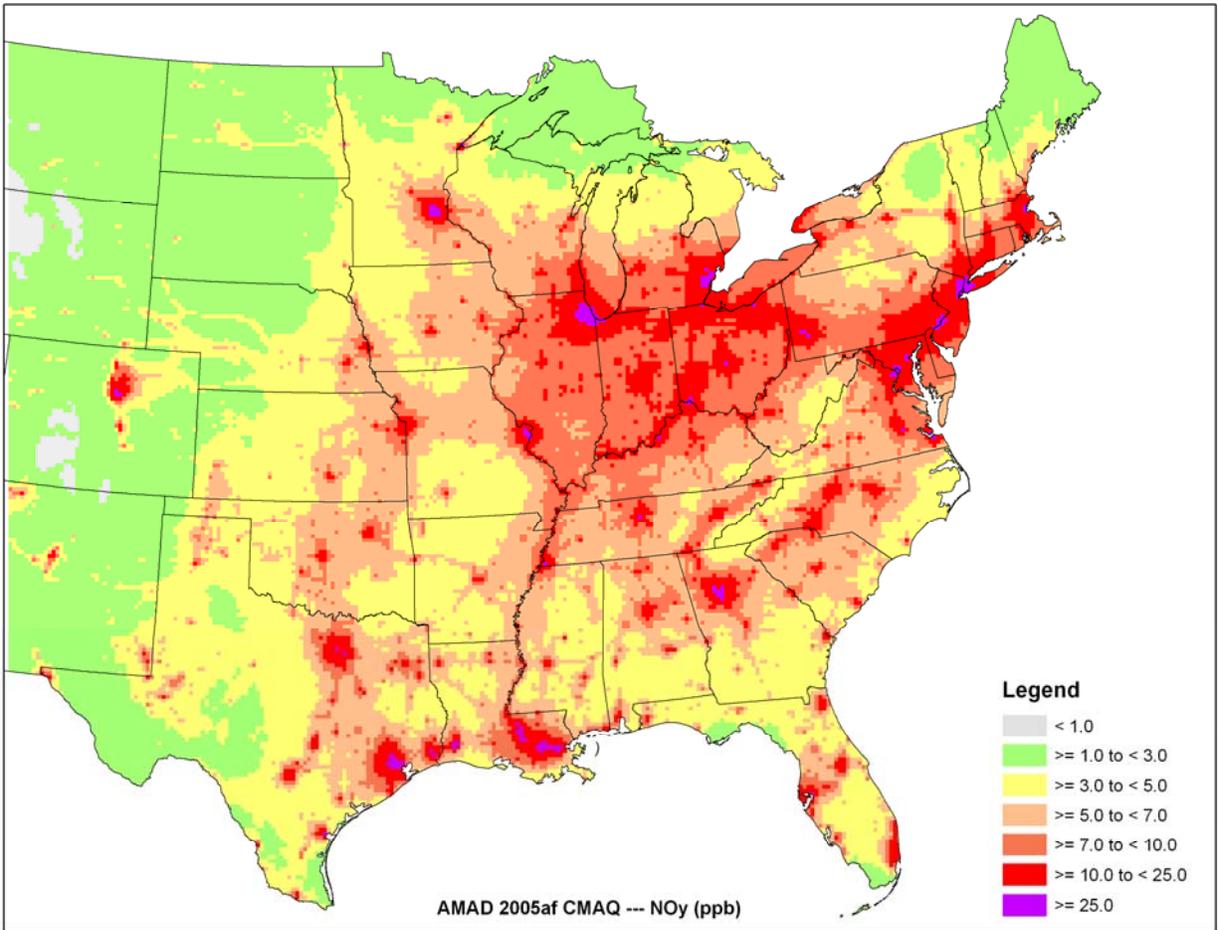
22 Nationwide maps of CMAQ-predicted 2005 annual average  $\text{NO}_Y$ ,  $\text{NH}_X$  ( $\text{NH}_3$  and  $\text{NH}_4$ ),  
23  $\text{NH}_3$ ,  $\text{NH}_4$ ,  $\text{SO}_X$ ,  $\text{SO}_4$ , and  $\text{SO}_2$  are provided in Figures 4-5 through 4-11 respectively. Given the  
24 considerable gaps in air quality observation networks as discussed in the REA and ISA (2008),  
25 modeled concentration patterns are used here to illustrate national representations of current air  
26 quality conditions for nitrogen and sulfur. The 2005 model year reflects the most recent  
27 available simulation for inclusion in this policy assessment. In addition, Figures 4-12 and 4-13  
28 provide maps of 2005 annual average  $\text{SO}_2$  and  $\text{SO}_4$ , respectively based on CASTNET  
29 observations. Site specific annual average 2005  $\text{NO}_Y$  measured concentrations at SLAMS  
30 (Figure 4-14) are typically are less than 40 ppb. The spatial patterns for the 2005 modeled and  
31 observed  $\text{NO}_Y$  and  $\text{SO}_2$  concentrations are similar to the 2002 CMAQ-based maps provided in

1 the REA, largely capturing the influence of major source regions throughout the nation. A  
2 spreading of the oxidized sulfur fields (Figures 4-5 and 4-6), relative to SO<sub>2</sub>, is consistent with  
3 sulfate transformation and associated air mass aging and transport. Ammonia and ammonium  
4 concentration patterns (Figure 4-4) are influenced strongly by the ammonia emissions  
5 distribution, with marginal spreading associated with the formation of NH<sub>4</sub>. The NH<sub>x</sub> fields are  
6 more strongly influenced by source location, relative to sulfur, based on the fast removal of  
7 atmospheric ammonia through deposition. However, recent incorporation of ammonia bi-  
8 directional flux treatment (see Chapter 7) does reduce NH<sub>3</sub> spatial gradients. Total deposition  
9 for nitrogen and sulfur (Figures 4-15 and 4-16) basically follow the patterns of ambient air  
10 concentrations. The contribution of reduced nitrogen to total nitrogen deposition (Figure 4-17)  
11 illustrates the strong influence of agricultural based ammonia emissions, particularly in upper  
12 midwest and eastern North Carolina.

13  
14 The 2005 ambient conditions indicate that the current SO<sub>2</sub> and NO<sub>2</sub> secondary standards  
15 are not exceeded (Figures 4-18 and 4-19) in locations where ecological effects have been  
16 observed, and where critical loads of nitrogen and sulfur are exceeded. This information is  
17 consistent with the fact that NO<sub>2</sub> accounts for only a fraction of NO<sub>y</sub>, and thus decreases in NO  
18 and NO<sub>2</sub> emissions that result in attaining the current secondary NO<sub>2</sub> standard would not be  
19 expected to fully address deposition of NO<sub>y</sub> in acid sensitive areas that generally are not  
20 represented by urban oriented NO<sub>2</sub> monitoring locations. The map in Figure 4-20 further  
21 illustrates this point by showing that the contribution of NO<sub>2</sub> to NO<sub>y</sub> is often less than 50% in  
22 rural areas. Neither NO<sub>y</sub> nor NO<sub>2</sub> concentrations correlate well with total oxidized nitrogen  
23 deposition (Figure 4-21), based on the annual average values in each 12 km CMAQ grid cell.  
24 The lack of correlation between NO<sub>y</sub> and nitrogen deposition is largely due to the inclusion of  
25 NO<sub>y</sub> species with low deposition velocity, primarily NO<sub>2</sub>. While NO<sub>2</sub> does not reflect the  
26 majority of oxidized nitrogen in rural environments, the species remains a significant contributor  
27 to the total ambient NO<sub>y</sub> budget. In contrast, species with high deposition velocities such as  
28 nitric acid correlate well with oxidized nitrogen deposition. The temporal correlations between  
29 NO<sub>y</sub> and deposition are likely to be similar to the nitric acid to deposition relationship when  
30 averaged over larger a spatial area as the influence of species with low deposition velocities is

1 minimized in rural locations associated with acid sensitive areas. Those relationships will be  
2 explored in the final PAD.

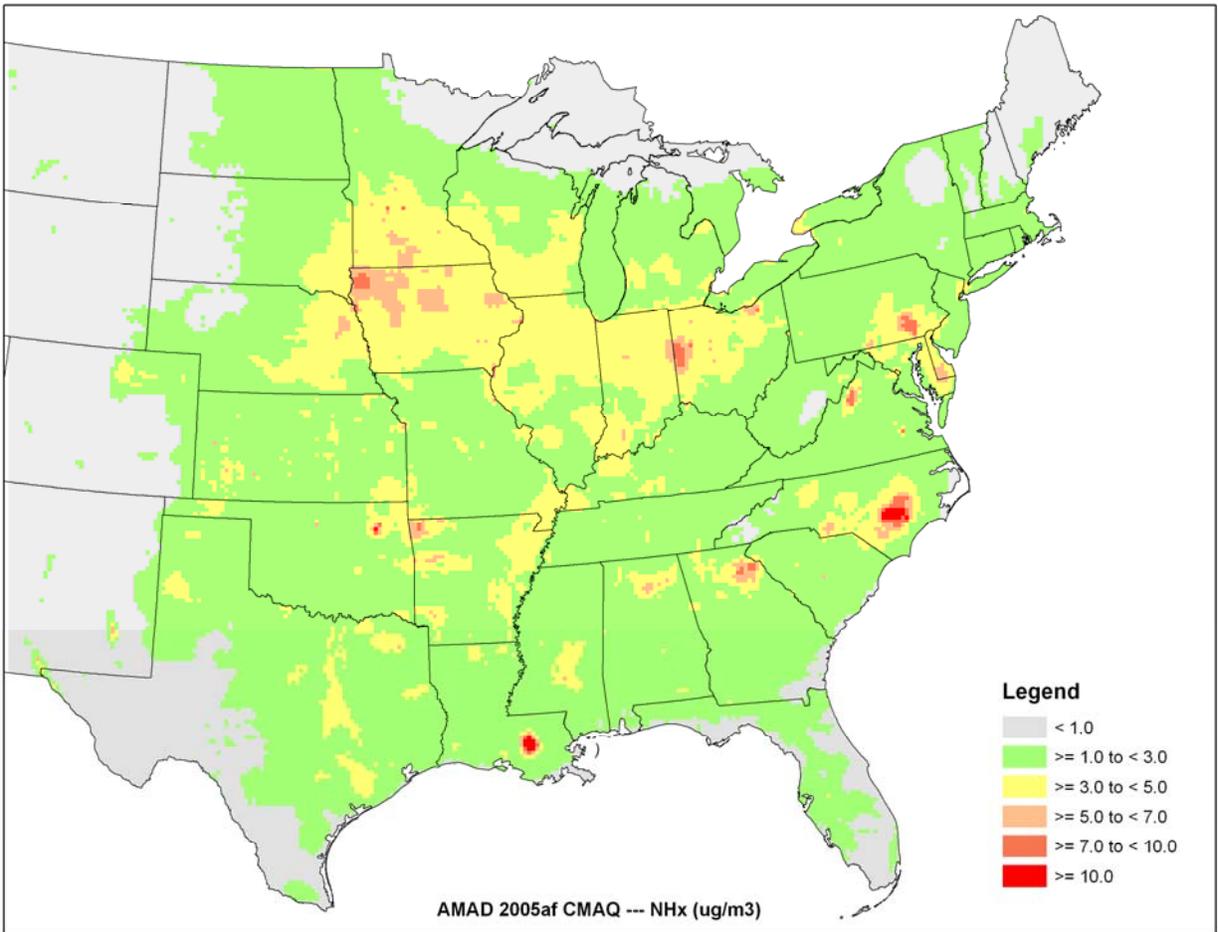
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**Figure 4-5. 2005 CMAQ modeled annual average NO<sub>y</sub> (ppb; see Table 1-1 for unit conversions).**

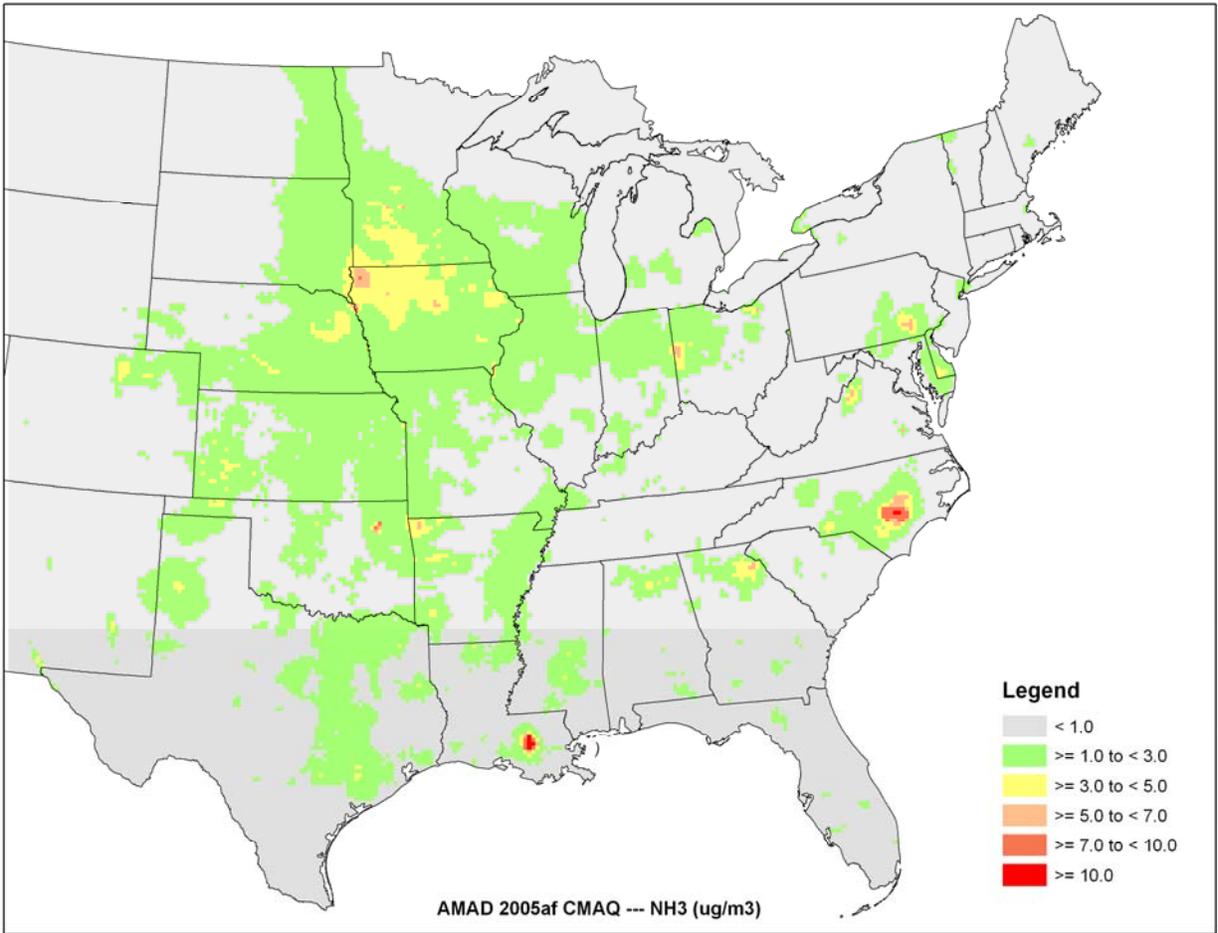
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**Figure 4-6. 2005 CMAQ modeled annual average total reduced nitrogen (NH<sub>x</sub>) (as ug/m<sup>3</sup> nitrogen – see Table 1-1 for unit conversions)**

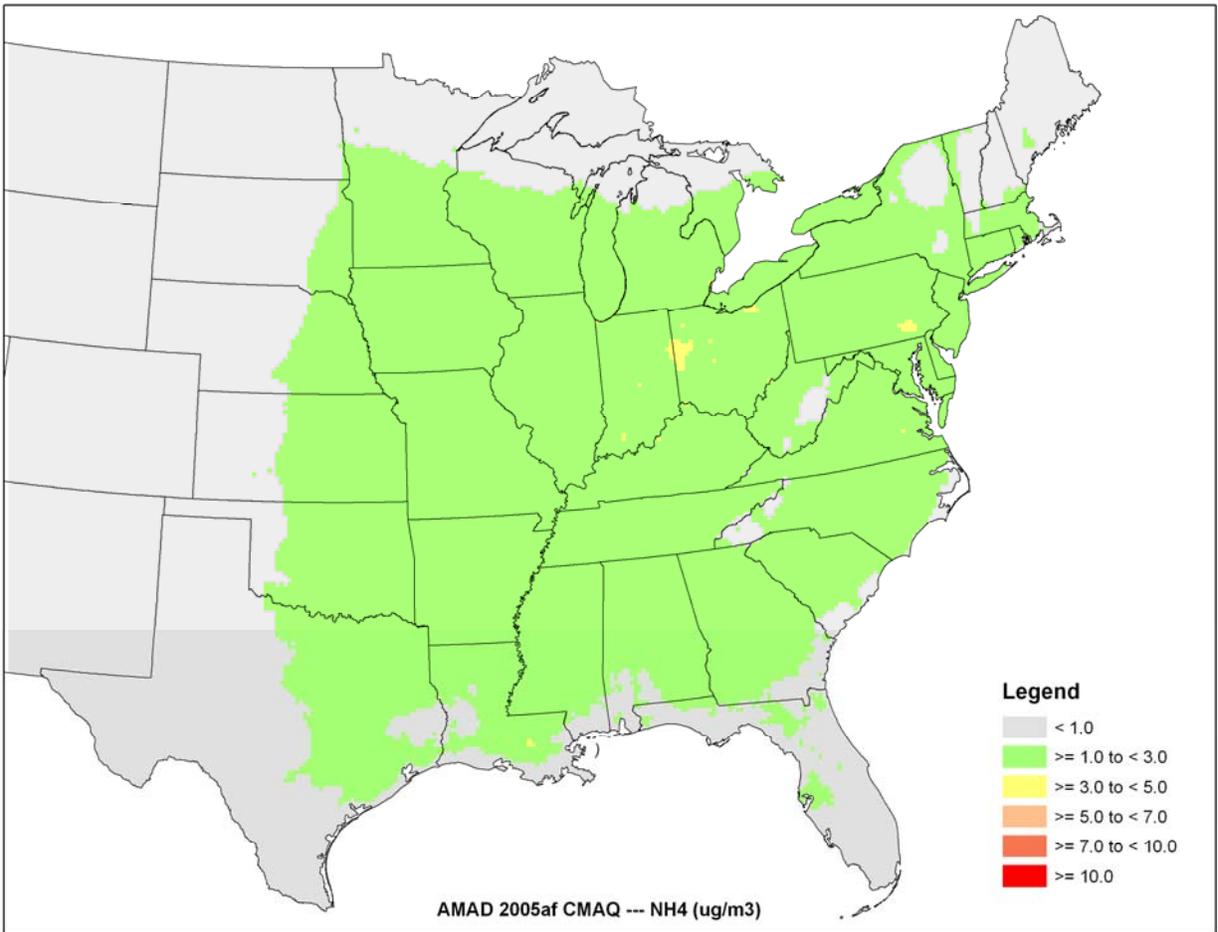
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**Figure 4-7. 2005 CMAQ modeled annual average ammonia, NH<sub>3</sub>, (as ug/m<sup>3</sup> N; see Table 1-1 for unit conversions )**

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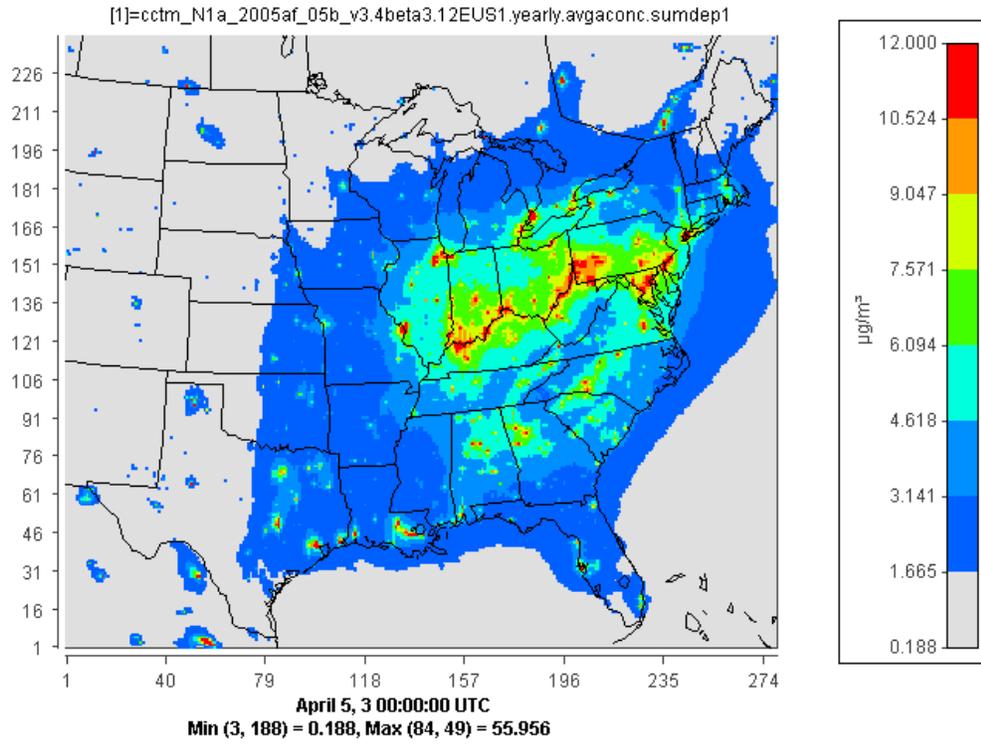
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**Figure 4-8. 2005 CMAQ modeled annual average ammonium, NH<sub>4</sub>, (as ug/m<sup>3</sup> N; see Table 1-1 for unit conversions)**

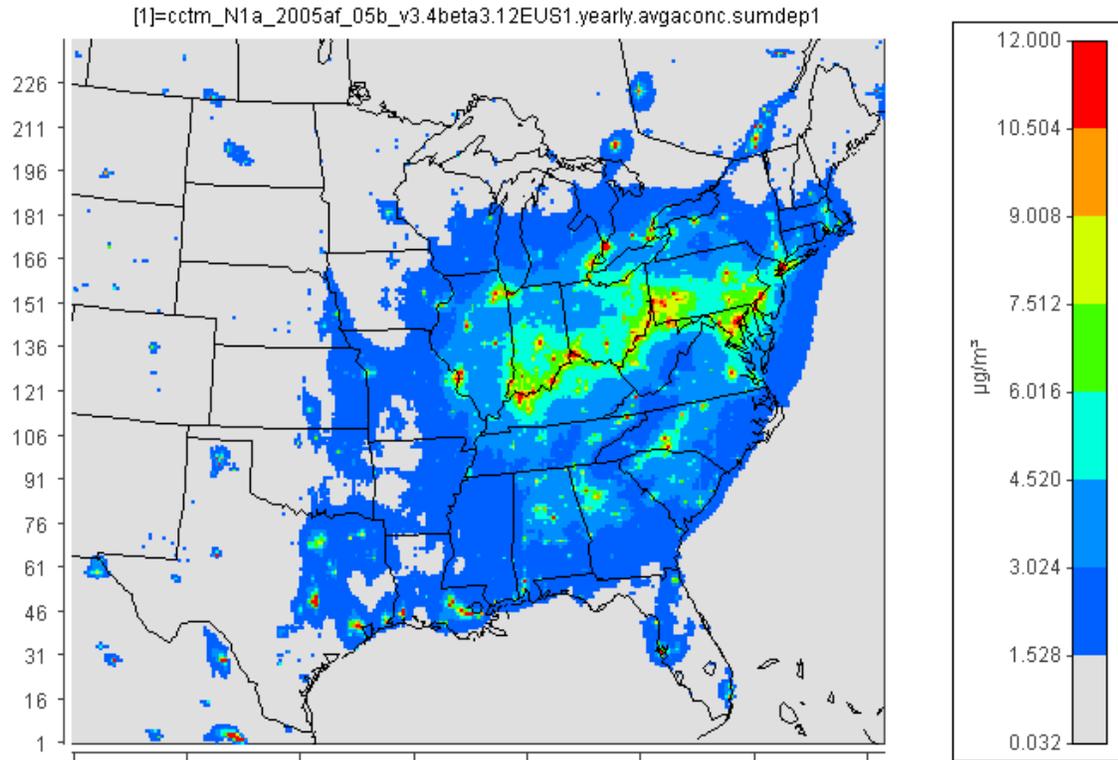
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**Figure 4-9. 2005 CMAQ modeled annual average SO<sub>x</sub>, (as ug/m<sup>3</sup> S from SO<sub>2</sub> and SO<sub>4</sub>; see Table 1-1 for unit conversions).**

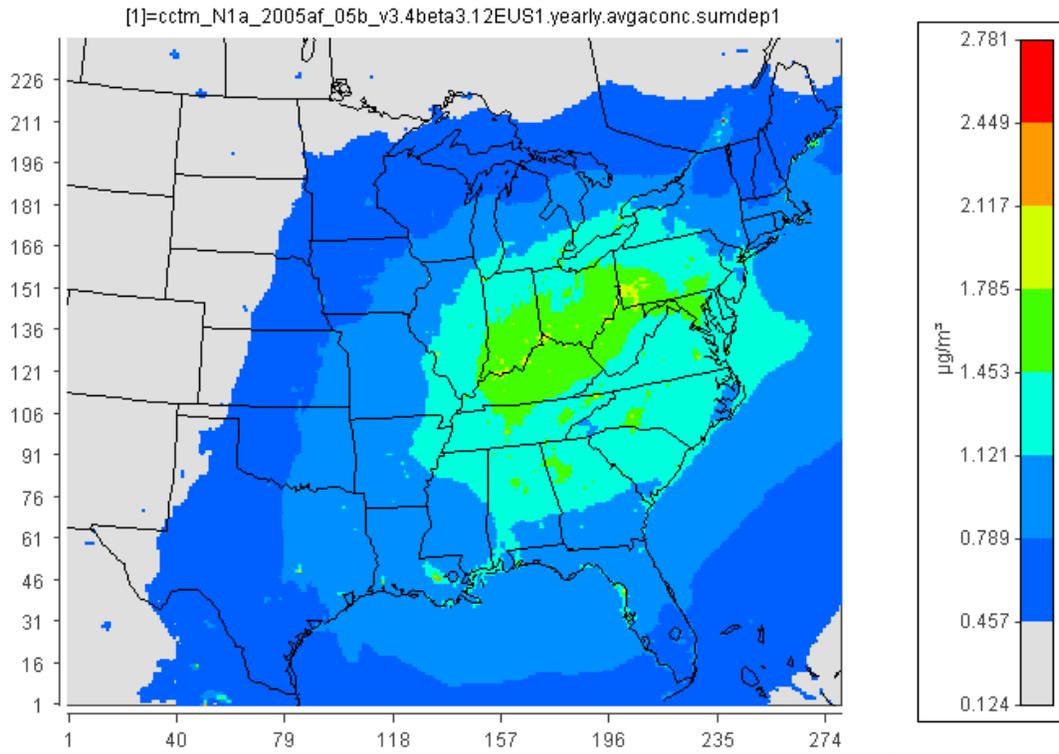
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**Figure 4-10. 2005 CMAQ modeled annual average SO<sub>2</sub> (as ug/m<sup>3</sup> S; see Table 1-1 for unit conversions )**

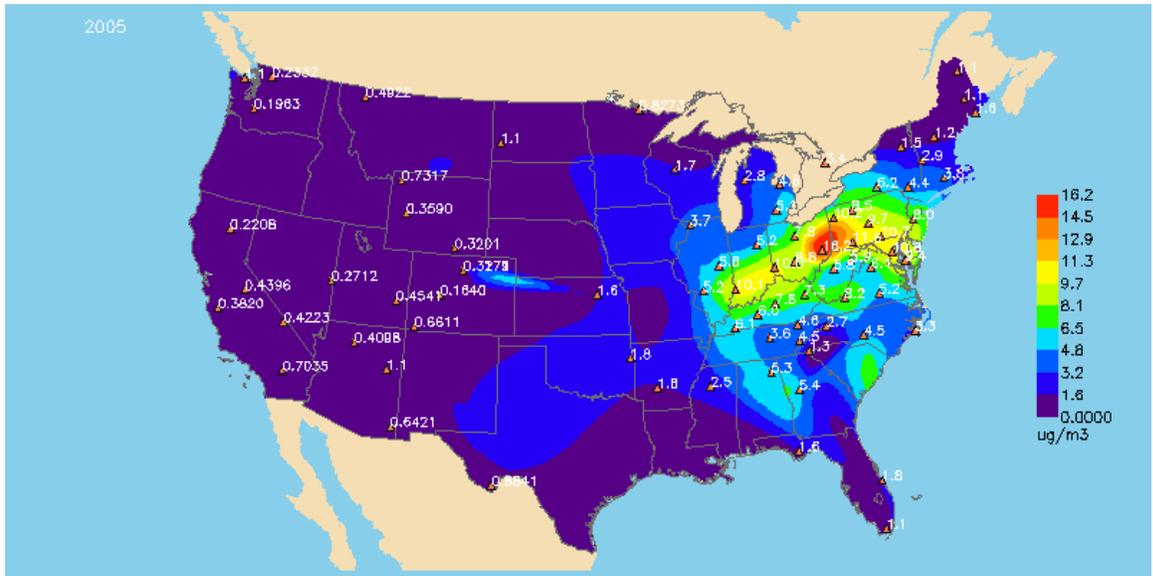
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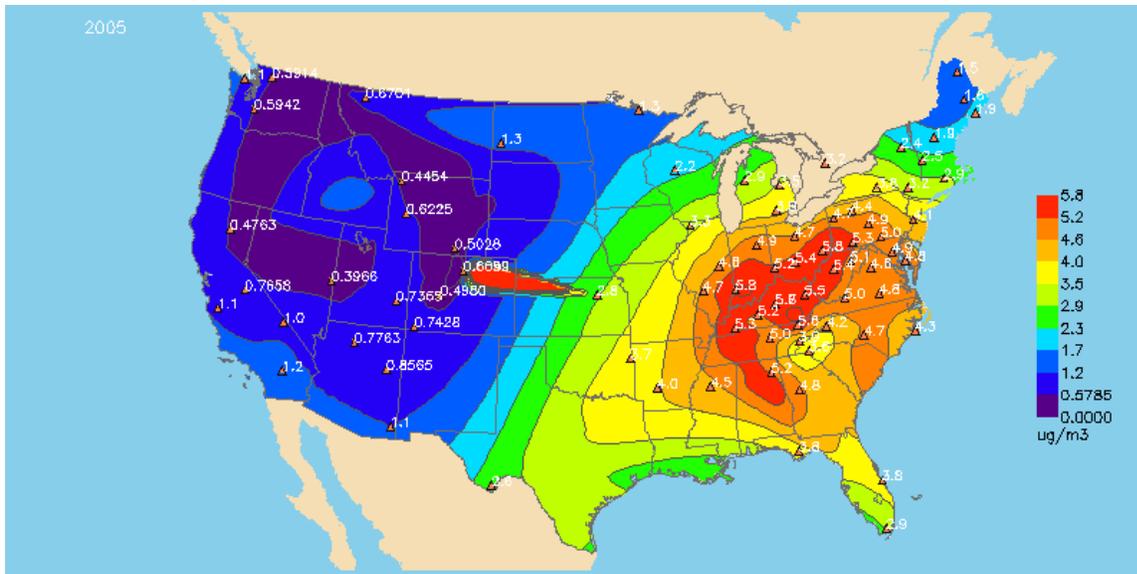
**Figure 4-11. 2005 CMAQ modeled annual average SO<sub>4</sub> (as ug/m<sup>3</sup> S; see Table 1-1 for unit conversions).**

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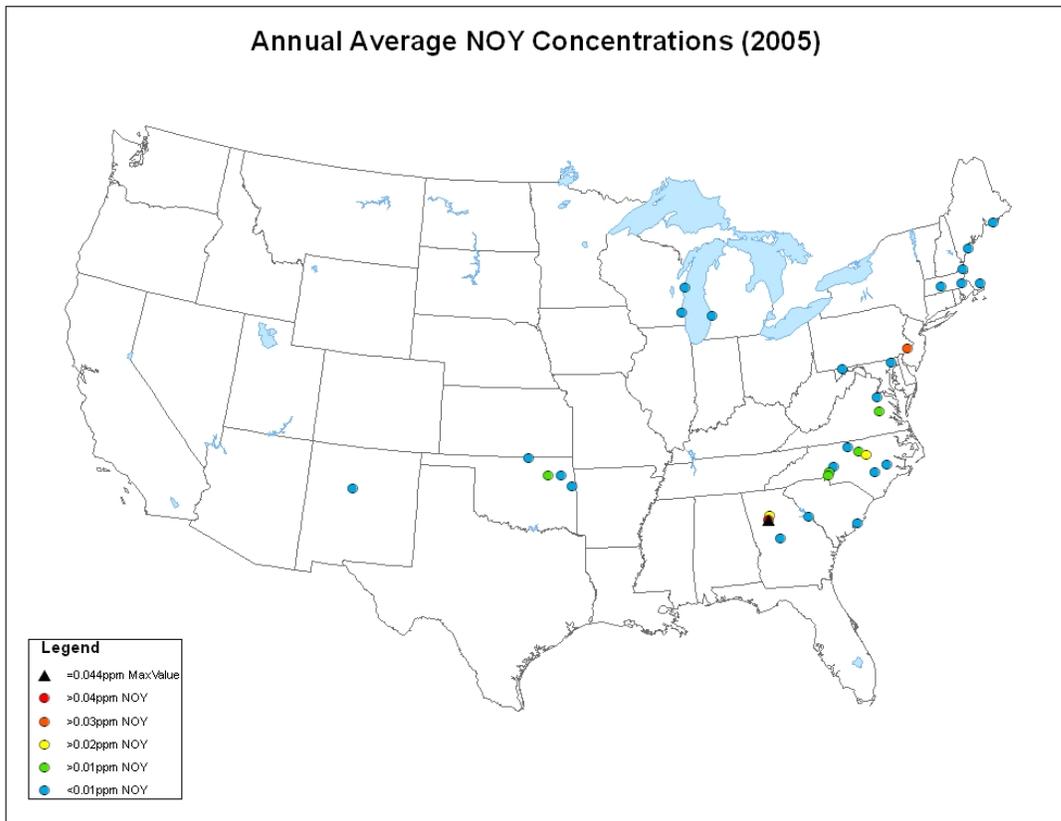
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**Figure 4-12. 2005 annual average sulfur dioxide concentrations based on CASTNET generated by the Visibility Information Exchange Web System (VIEWS). See Table 1-1 for unit conversions**



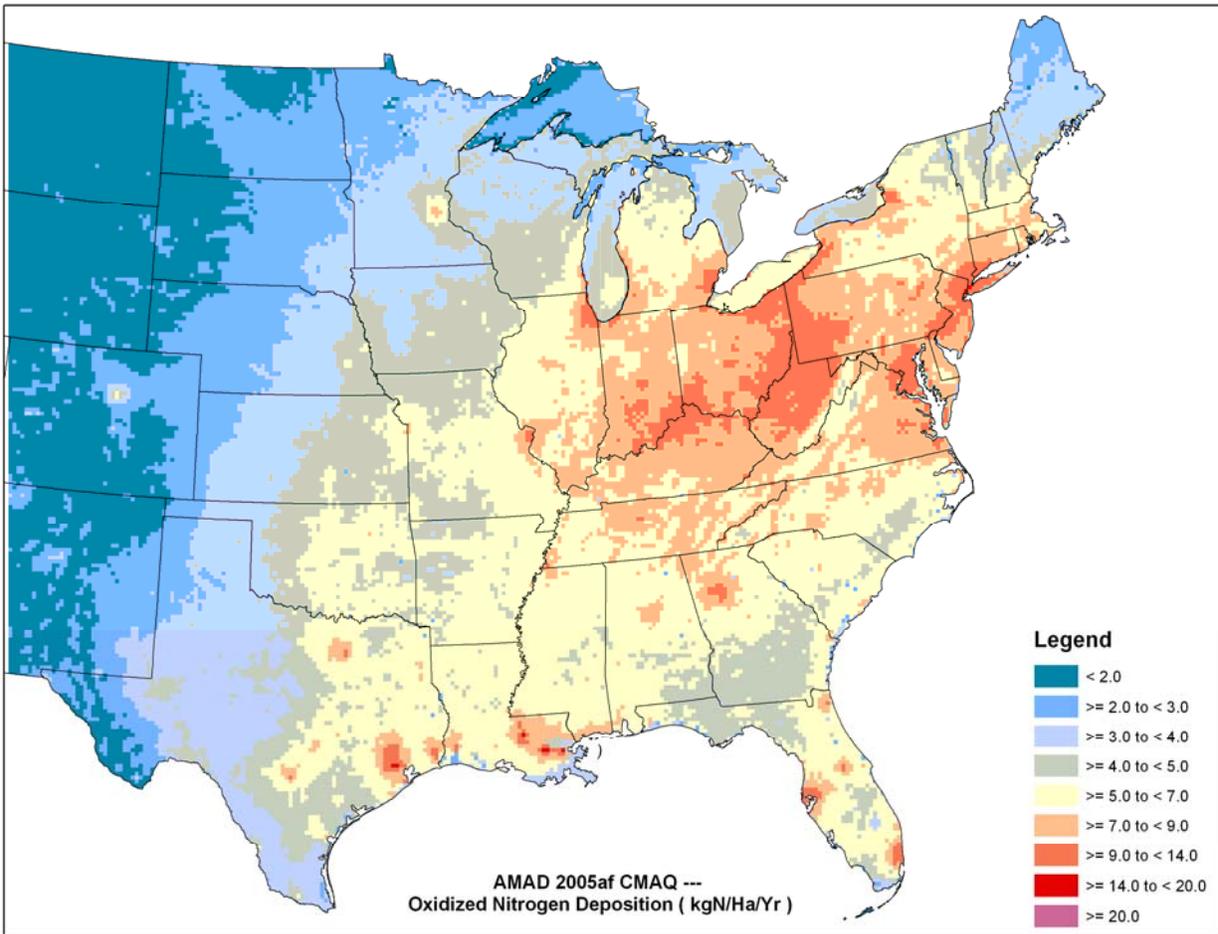
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**Figure 4-13** 2005 annual average sulfate concentrations based on CASTNET generated by the Visibility Information Exchange Web System (VIEWS). [interpolating relative sparse data can produce unrealistic concentration plumes as demonstrated in the central U.S.] see Table 1-1 for unit conversions.



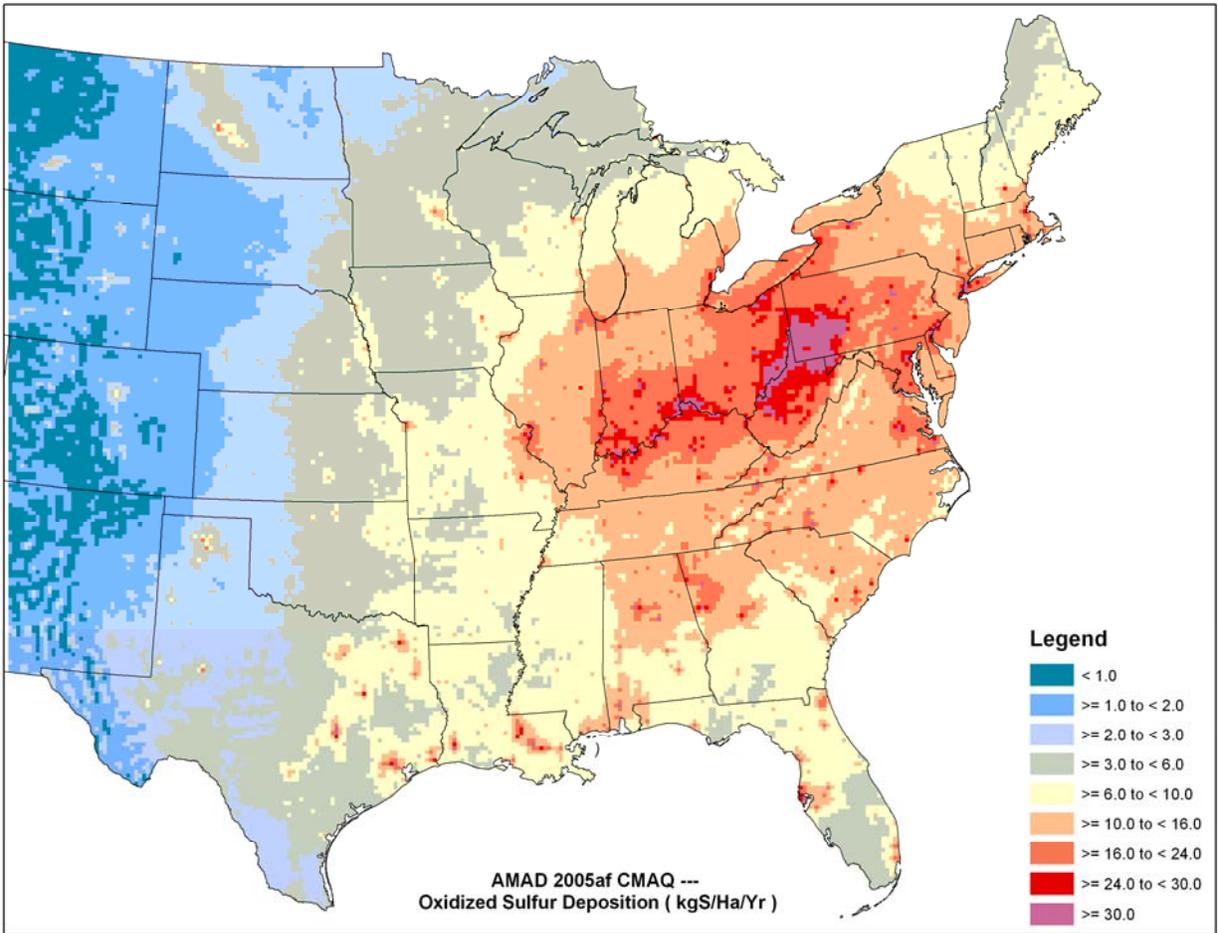
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 2 **Figure 4-14. Annual average 2005 NO<sub>y</sub> concentrations from reporting stations in the Air**  
 3 **Quality System (AQS). see Table 1-1 for unit conversions.**

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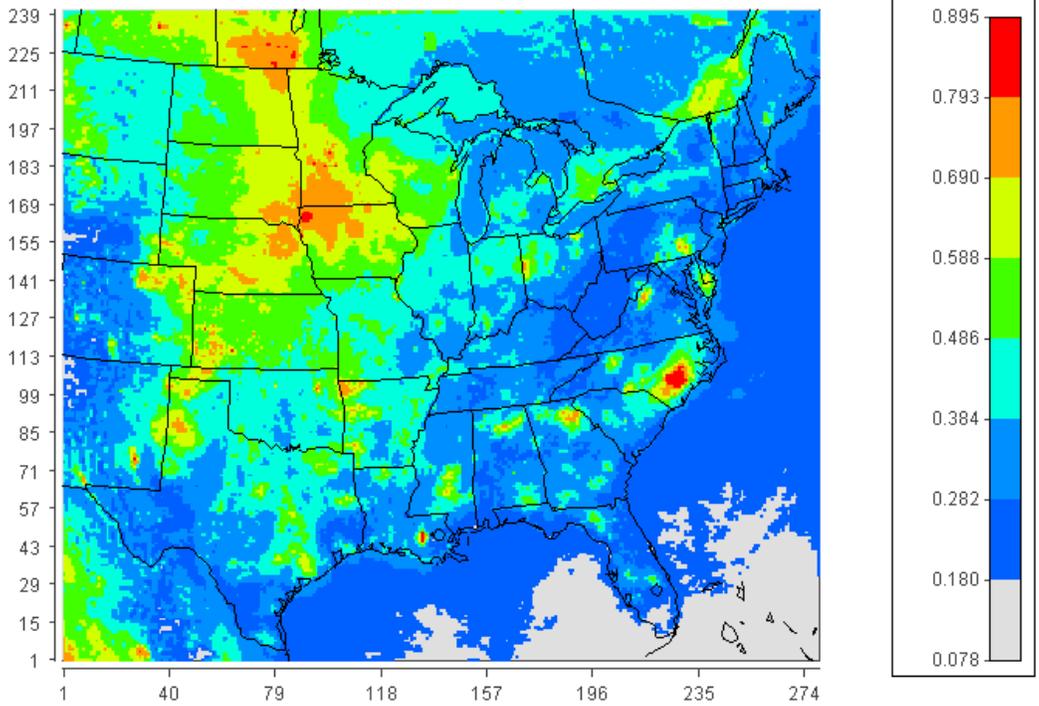
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**Figure 4-15. 2005 CMAQ modeled oxidized nitrogen deposition (kgN/ha-yr).** see Table 1-1 for unit conversions



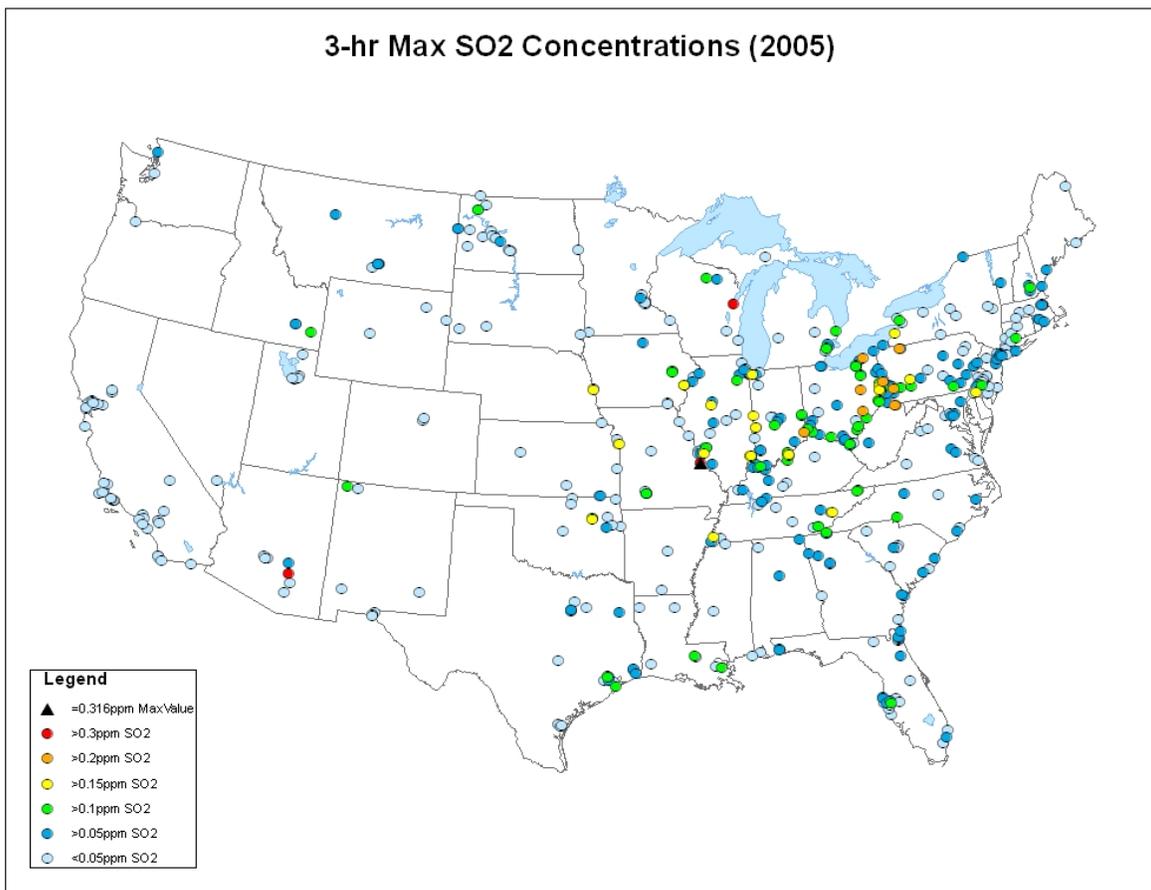
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2 **Figure 4-16. 2005 CMAQ modeled oxidized sulfur deposition (kgS/ha-yr).** see Table 1-1  
3 for unit conversions

### Fraction of Reduced to Total N Deposition

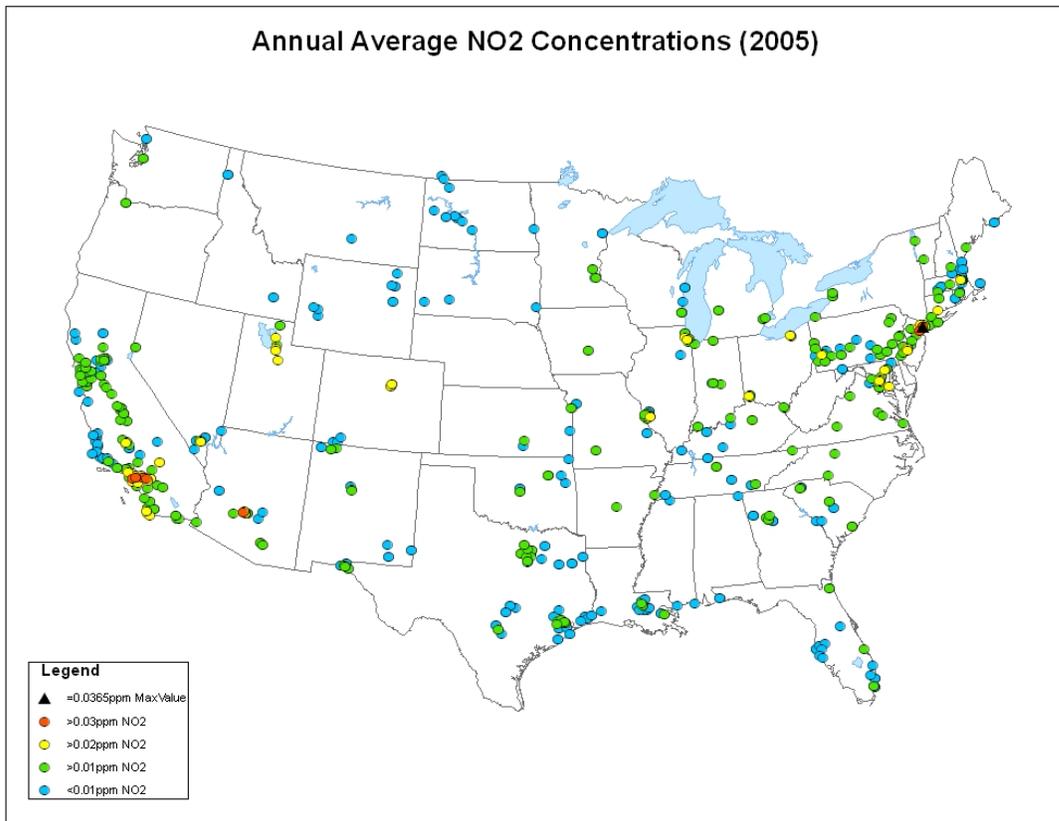


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**Figure 4-17. 2005 CMAQ derived annual average ratio of reduced to total nitrogen deposition.**

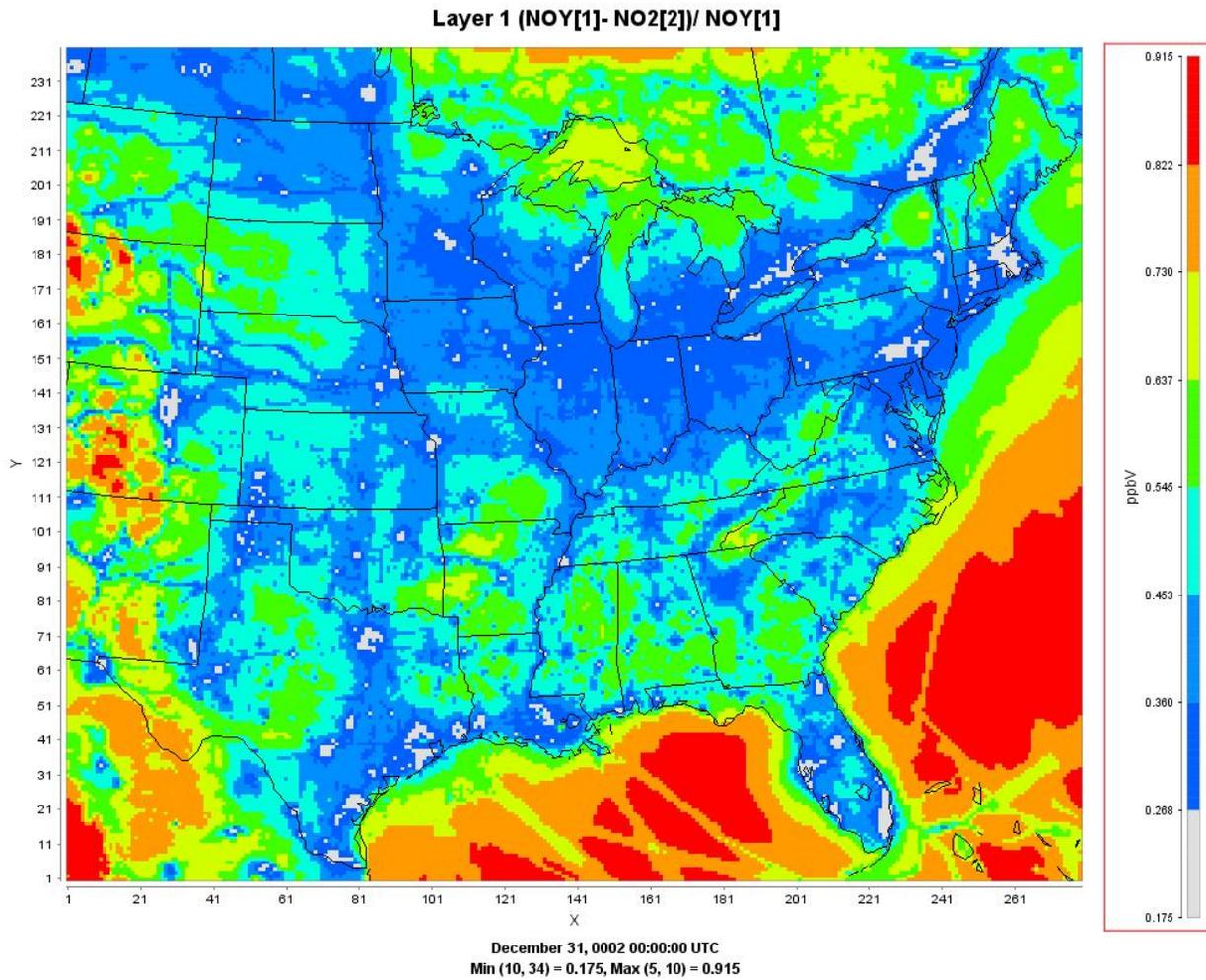


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3 **Figure 4-18. Three hour average maximum 2005 SO<sub>2</sub> concentrations based on the SLAMS**  
4 **reporting to EPA's Air Quality System (AQS) data base.** The current SO<sub>2</sub>  
5 secondary standard based on the maximum 3 hour average value is 500 ppb, a  
6 value not exceeded. While there are obvious spatial gaps, the majority of these  
7 stations are located to capture maximum values generally in proximity to major  
8 sources and high populations. Lower relative values are expected in more remote  
9 acid sensitive areas. see Table 1-1 for unit conversions



1  
 2 **Figure 4-19 Annual average 2005 NO<sub>2</sub> concentrations based on the SLAMS reporting to**  
 3 **EPA's Air Quality System (AQS) data base.** The current NO<sub>2</sub> secondary  
 4 standard is 53 ppb, a value well above those observed. While there are obvious  
 5 spatial gaps, the stations are located in areas of relatively high concentrations in  
 6 highly populated areas. Lower relative values are expected in more remote acid  
 7 sensitive areas. see Table 1-1 for unit conversions

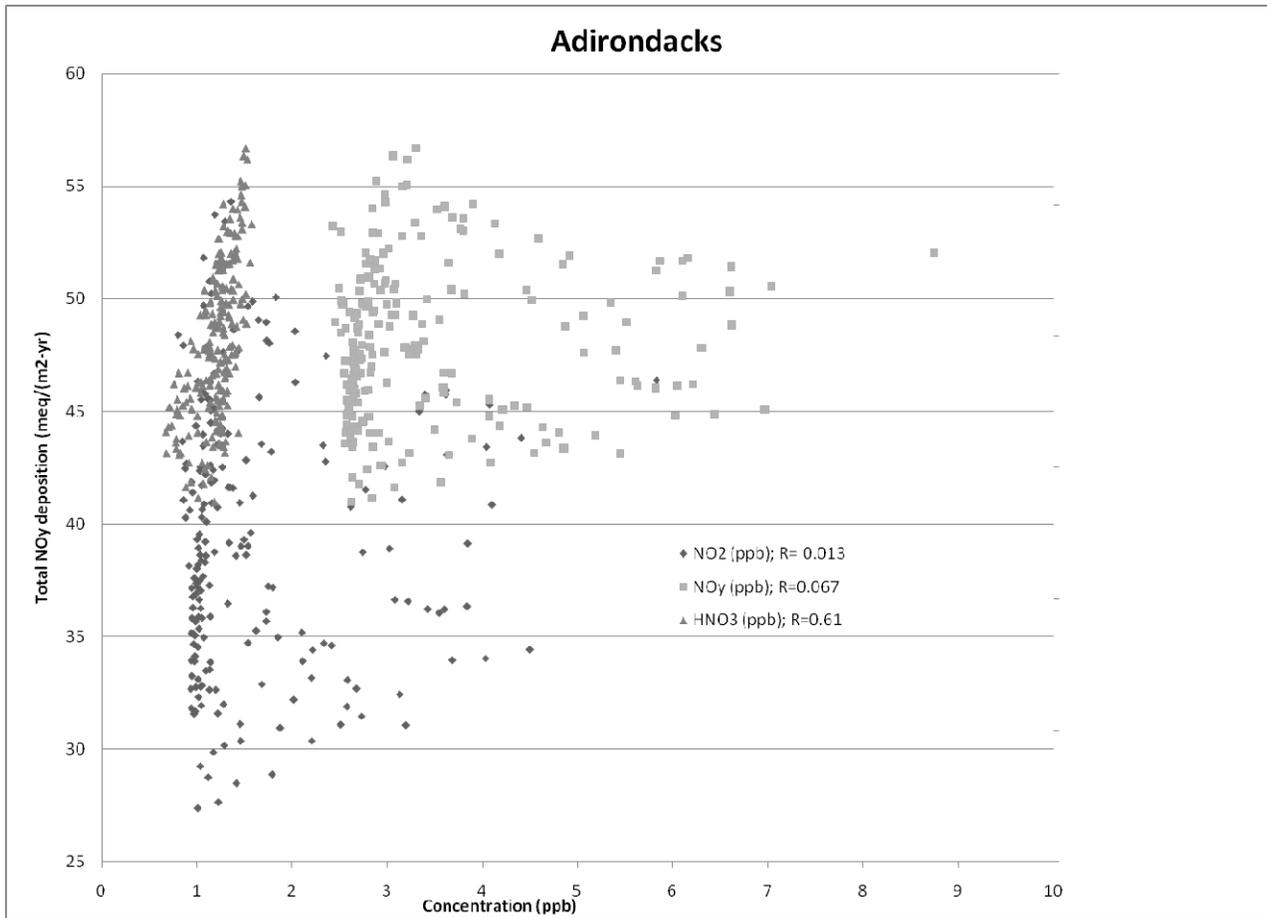
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**Figure 4-20** 2005 CMAQ derived annual average ratio of  $(\text{NO}_Y - \text{NO}_2)/\text{NO}_Y$ . The fraction of  $\text{NO}_2$  contributing to total  $\text{NO}_Y$  generally is less than 50% in the Adirondack and Shenandoah case study areas. The ratio (dimensionless – scale units of ppbv are an automated output) reflects the relative air mass aging associated with transformation of oxidized nitrogen beyond  $\text{NO}$  and  $\text{NO}_2$  as one moves from urban to rural locations.

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**Figure 4-21** Scatter plots of total oxidized nitrogen deposition with average annual NO<sub>2</sub>, HNO<sub>3</sub> and NO<sub>y</sub> concentrations on each 12 km<sup>2</sup> grid based on 2005 CMAQ results for the Adirondack region.

1  
2 **4.5 Are Adverse Effects On The Public Welfare Occurring Under Current Air Quality**  
3 **Conditions For NO<sub>2</sub> And SO<sub>2</sub> And Would They Occur If The Nation Met The**  
4 **Current Secondary Standards?**  
5

6 In the previous sections we have established that almost all areas of the U.S. were at  
7 concentrations of SO<sub>2</sub> and NO<sub>2</sub> below the levels of the current standards. In many locations, SO<sub>2</sub>  
8 and NO<sub>2</sub> concentrations are substantially below the levels of the standards. This pattern suggests  
9 that levels of deposition and any effects on ecosystems due to deposition of NO<sub>x</sub> and SO<sub>x</sub> under  
10 recent conditions are occurring even though areas meet or are below current standards. In this  
11 section we focus on summarizing the evidence of effects occurring at deposition levels consistent  
12 with recent conditions.

13 The ISA summarizes the available studies of relative nitrogen contribution and finds that  
14 in much of the U.S., NO<sub>x</sub> contributes from 50 to 75 percent of total atmospheric deposition  
15 relative to total reactive nitrogen that includes oxidized and reduced nitrogen species.[**ISA**  
16 **Section 2.8.4**]. Although the proportion of total nitrogen loadings associated with atmospheric  
17 deposition of nitrogen varies across locations (N deposition in the eastern U.S. includes locations  
18 with greater than 9 kg N/ha-yr, and in the central U.S. high deposition locations with values on  
19 the order of 6 to 7 kg N/ha-yr), the ISA indicates that atmospheric N deposition is the main  
20 source of new anthropogenic N to most headwater streams, high elevation lakes, and low-order  
21 streams. Atmospheric N deposition contributes to the total N load in terrestrial, wetland,  
22 freshwater, and estuarine ecosystems that receive N through multiple pathways. In several large  
23 estuarine systems, including the Chesapeake Bay, atmospheric deposition accounts for between  
24 10 and 40 percent of total nitrogen loadings (U.S. EPA, 2000).

25 Atmospheric concentrations of SO<sub>x</sub> account for nearly all S deposition in the US. For the  
26 period 2004–2006, mean S deposition in the U.S. was greatest east of the Mississippi River with  
27 the highest deposition amount, 21.3 kg S/ha-yr, in the Ohio River Valley where most recording  
28 stations reported 3 year averages >10 kg S/ha-yr. Numerous other stations in the East reported S  
29 deposition >5 kg S/ha-yr. Total S deposition in the U.S. west of the 100th meridian was  
30 relatively low, with all recording stations reporting <2 kg S/ha-yr and many reporting <1 kg

1 S/ha-yr. S was primarily deposited in the form of wet  $\text{SO}_4^{2-}$  followed in decreasing order by a  
2 smaller proportion of dry  $\text{SO}_2$  and a much smaller proportion of deposition as dry  $\text{SO}_4^{2-}$ .

3 New scientific evidence exists to address each of the areas of uncertainty raised in the  
4 previous reviews (summarized above in section 1.4). Based on the new evidence, the current  
5 ISA concludes that:

6 (1) The evidence is sufficient to infer a causal relationship between acidifying  
7 deposition (to which both  $\text{NO}_x$  and  $\text{SO}_x$  contribute) and effects on  
8 biogeochemistry related to terrestrial and aquatic ecosystems; and biota in  
9 terrestrial and aquatic ecosystems.

10 (2) The evidence is sufficient to infer a causal relationship between N deposition,  
11 to which  $\text{NO}_x$  and  $\text{NH}_x$  contribute, and the alteration of A) biogeochemical  
12 cycling of N and carbon in terrestrial, wetland, freshwater aquatic, and coastal  
13 marine ecosystems; B) biogenic flux of methane ( $\text{CH}_4$ ), and  $\text{N}_2\text{O}$  in terrestrial  
14 and wetland ecosystems; and C) species richness, species composition, and  
15 biodiversity in terrestrial, wetland, freshwater aquatic and coastal marine  
16 ecosystems.

17 (3) The evidence is sufficient to infer a causal relationship between S deposition  
18 and increased Hg methylation in wetlands and aquatic environments.

19 Subsequent to the previous review of the  $\text{NO}_x$  secondary standard, a great deal of  
20 information on the contribution of atmospheric deposition associated with ambient  $\text{NO}_x$  has  
21 become available. In Chapter 3 of the REA a thorough assessment is provided of the  
22 contribution of  $\text{NO}_x$  to nitrogen deposition throughout the U.S., and the relative contributions of  
23 ambient  $\text{NO}_x$  and reduced forms of nitrogen. Staff concludes that based on that analysis,  
24 ambient  $\text{NO}_x$  is a significant component of atmospheric nitrogen deposition, even in areas with  
25 relatively high rates of deposition of reduced nitrogen. In addition, staff concludes that  
26 atmospheric deposition of oxidized nitrogen contributes significantly to total nitrogen loadings in  
27 nitrogen sensitive ecosystems.

28 As discussed throughout the REA document, there are several key areas of risk that are  
29 associated with ambient concentrations of  $\text{NO}_x$  and  $\text{SO}_x$ . As noted earlier, in previous reviews  
30 of the  $\text{NO}_x$  and  $\text{SO}_x$  secondary standards, the standards were designed to protect against direct  
31 exposure of plants to ambient concentrations of the pollutants. A significant shift in

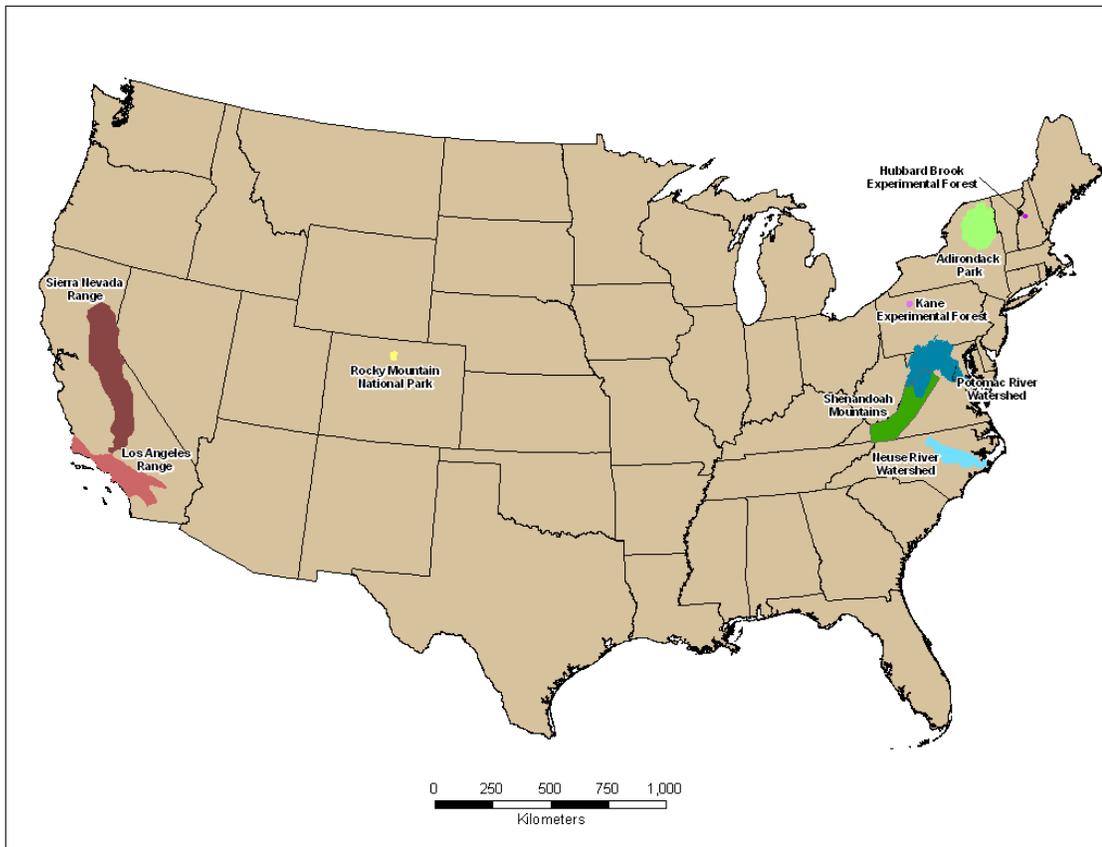
1 understanding of the effects of NO<sub>x</sub> and SO<sub>x</sub> has occurred since the last reviews, reflecting the  
2 large amount of research that has been conducted on the effects of deposition of nitrogen and  
3 sulfur to ecosystems. The most significant risks of adverse effects to public welfare are those  
4 related to deposition of NO<sub>x</sub> and SO<sub>x</sub> to both terrestrial and aquatic ecosystems. These risks fall  
5 into two categories: acidification and nutrient enrichment. These made up the emphasis of the  
6 REA, and are most relevant to evaluating the adequacy of the existing standards in protecting  
7 public welfare from adverse ecological effects.

8

9 **4.5.1 To what extent do the current NO<sub>x</sub> and SO<sub>x</sub> secondary standards provide**  
10 **protection from adverse effects associated with deposition of atmospheric NO<sub>x</sub> and SO<sub>x</sub>**  
11 **which results in acidification in sensitive aquatic and terrestrial ecosystems?**

12

13 The focus of the REA case studies was on determining whether deposition of sulfur and  
14 oxidized nitrogen in locations where ambient NO<sub>x</sub> and SO<sub>x</sub> was at or below the current  
15 standards was resulting in acidification and related effects. This review has focused on  
16 identifying ecological indicators that can link atmospheric deposition to ecological effects  
17 associated with acidification. NO<sub>x</sub> and SO<sub>x</sub> contribute to acidification in both aquatic and  
18 terrestrial ecosystems, although the indicators of effects differ. Although there are some  
19 geographic areas with both terrestrial and aquatic ecosystems that are vulnerable to acidification,  
20 the case study areas do not fully overlap. The locations of the case studies evaluated in the REA  
21 are shown on Figure 4-20.



1  
 2 **Figure 4-22 National map highlighting the nine case study areas evaluated in the REA.**

3 **4.5.1.1 Aquatic Acidification**

4 Based on the case studies conducted for lakes in the Adirondacks and streams in  
 5 Shenandoah National Park, staff concludes that there is significant risk to acid sensitive aquatic  
 6 ecosystems at atmospheric concentrations of  $\text{NO}_x$  and  $\text{SO}_x$  at or below the current standards.  
 7 This conclusion is based on application of the MAGIC model to estimate the effects of  
 8 deposition at levels consistent with atmospheric  $\text{NO}_x$  and  $\text{SO}_x$  concentrations that are at or  
 9 below the current standards. An important ecological indicator for aquatic acidification effects is  
 10 acid neutralizing capacity (ANC) of a waterbody, and the case study focused on evaluating  
 11 whether locations were likely to be below critical values of ANC given deposition levels  
 12 associated with  $\text{NO}_x$  and  $\text{SO}_x$  atmospheric concentrations that meet the current standards. In  
 13 addition, the case studies assessed the ecological effects and some of the known ecosystem  
 14 services that are associated with different levels of ANC in order to associate levels of ANC with

1 measures of public welfare that may be adversely affected by deposition levels consistent with  
2 atmospheric concentrations of NO<sub>x</sub> and SO<sub>x</sub> that meet the current standards.

3 Staff concludes that the evidence and risk assessment support strongly a relationship  
4 between atmospheric deposition of NO<sub>x</sub> and SO<sub>x</sub> and loss of ANC in sensitive ecosystems, and  
5 that ANC is an excellent indicator of aquatic acidification. Staff also concludes that at levels of  
6 deposition associated with NO<sub>x</sub> and SO<sub>x</sub> concentrations at or below the current standards, ANC  
7 levels are expected to be below benchmark values that are associated with significant losses in  
8 fish species richness (**REA Section 4**).

9 Many locations in sensitive areas of the U.S. have ANC levels below benchmark levels  
10 for ANC classified as severe, elevated, or moderate concern (see Figure 2-1). The average  
11 current ANC levels across 44 lakes in the Adirondack case study area is 62.1 µeq/L (moderate  
12 concern). However, 44 percent of lakes had deposition levels exceeding the critical load for an  
13 ANC of 50 µeq/L, and 28 percent of lakes had deposition levels exceeding the critical load for an  
14 ANC of 20 µeq/L (**REA Section 4.2.4.2**). This information indicates that almost half of the 44  
15 lakes in the Adirondacks case study area are at an elevated concern levels, and almost a third are  
16 at a severe concern level. These levels are associated with greatly diminished fish species  
17 diversity, and losses in the health and reproductive capacity of remaining populations. Based on  
18 assessments of the relationship between number of fish species and ANC level in both the  
19 Adirondacks and Shenandoah areas, the number of fish species is decreased by over half at an  
20 ANC level of 20 µeq/L relative to an ANC level at 100 µeq/L (**REA Figure 4.2-1**). At levels  
21 below 20 µeq/L, populations of sensitive species, such as brook trout, may decline significantly  
22 during episodic acidification events. When extrapolated to the full population of lakes in the  
23 Adirondacks area using weights based on the EMAP probability survey (**REA 4.2.6.1**), 36  
24 percent of lakes exceeded the critical load for an ANC of 50 µeq/L and 13 percent of lakes  
25 exceeded the critical load for an ANC of 20 µeq/L.

26 Many streams in the Shenandoah case study area also have levels of deposition that are  
27 associated with ANC levels classified as severe, elevated, or moderate concern. The average  
28 ANC under recent conditions for the 60 streams evaluated in the Shenandoah case study area is  
29 57.9 µeq/L, indicating moderate concern. However, 85 percent of streams had recent deposition  
30 exceeding the critical load for an ANC of 50 µeq/L, and 72 percent exceeded the critical load for  
31 an ANC of 20 µeq/L. As with the Adirondacks area, this information suggests that significant

1 numbers of sensitive streams in the Shenandoah area are at risk of adverse impacts on fish  
2 populations under recent conditions. Many other streams in the Shenandoah area are likely to  
3 experience conditions of elevated to severe concern based on the prevalence in the area of  
4 bedrock geology associated with increased sensitivity to acidification suggesting that effects due  
5 to stream acidification could be widespread in the Shenandoah area (**REA 4.2.6.2**).

6 In the ISA it is noted that significant portions of the U.S. are acid sensitive, and that  
7 current deposition levels exceed those that would allow recovery of the most acid sensitive lakes  
8 in the Adirondacks (**ISA ES**). In addition, because of past loadings, areas of the Shenandoah are  
9 sensitive to current deposition levels (**ISA ES**). Parts of the West are naturally less sensitive to  
10 acidification and subjected to lower deposition (particularly SO<sub>x</sub>) levels relative to the eastern  
11 United States, and as such, less focus in the ISA is placed on the adequacy of the existing  
12 standards in these areas, with the exception of the mountainous areas of the West, which  
13 experience episodic acidification due to deposition.

14 While most (99 percent) of stream kilometers in the U.S. are not chronically acidified  
15 under current conditions, a recent survey found sensitive streams in many locations in the U.S.,  
16 including the Appalachian mountains, the Coastal Plain, and the Mountainous West (**ISA**  
17 **Section 4.2.2.3**). In these sensitive areas, between 1 and 6 percent of stream kilometers are  
18 chronically acidified.

19 The ISA notes that “consideration of episodic acidification greatly increases the extent  
20 and degree of estimated effects for acidifying deposition on surface waters.” (**ISA Section**  
21 **3.2.1.6**) Some studies show that the number of lakes that could be classified as acid-impacted  
22 based on episodic acidification is 2 to 3 times the number of lakes classified as acid-impacted  
23 based on chronic ANC. These episodic acidification events can have long term effects on fish  
24 populations (**ISA Section 3.2.1.6**). Under recent conditions, episodic acidification has been  
25 observed in locations in the eastern U.S. and in the mountainous western U.S. (**ISA Section**  
26 **3.2.1.6**).

27 It can therefore be concluded that recent levels of NO<sub>x</sub> and SO<sub>x</sub> are associated with  
28 deposition that leads to ANC values below benchmark values known to cause ecological harm in  
29 sensitive aquatic systems, including lakes and streams in multiple areas of the U.S. These  
30 changes are known to have impacts on ecosystem services including recreational fishing which is  
31 discussed along with other services in Chapter 3. While other ecosystem services (e.g. habitat

1 provisioning, subsistence fishing, and biological control as well as many others) are potentially  
2 affected by reductions in ANC, confidence in the specific translation of ANC values to these  
3 additional ecosystem services is much lower.

#### 4 5 **4.5.1.2 Terrestrial Acidification**

6 Based on the case studies on sugar maple and red spruce habitat, staff concludes that  
7 there is significant risk to sensitive terrestrial ecosystems from acidification at atmospheric  
8 concentrations of NO<sub>x</sub> and SO<sub>x</sub> at or below the current standards. This conclusion is based on  
9 application of the simple mass balance model to deposition levels associated with NO<sub>x</sub> and SO<sub>x</sub>  
10 concentrations at or below the current standards. The ecological indicator selected for terrestrial  
11 acidification is the base cation to aluminum ratio (BC:Al), which has been linked to tree health  
12 and growth. The results of the REA strongly support a relationship between atmospheric  
13 deposition of NO<sub>x</sub> and SO<sub>x</sub> and BC:Al, and that BC:Al is a good indicator of terrestrial  
14 acidification. At levels of deposition associated with NO<sub>x</sub> and SO<sub>x</sub> concentrations at or below  
15 the current standards, BC:Al levels are expected to be below benchmark values that are  
16 associated with significant effects on tree health and growth. Such degradation of terrestrial  
17 ecosystems could affect ecosystem services such as habitat provisioning, endangered species,  
18 goods production (timber, syrup, etc.) and many others.

19 Many locations in sensitive areas of the U.S. have BC:Al levels below benchmark levels  
20 classified as providing low to intermediate levels of protection to tree health. At a BC:Al ratio of  
21 1.2 (intermediate level of protection), red spruce growth can be reduced by 20 percent. At a  
22 BC:Al ratio of 0.6 (low level of protection), sugar maple growth can be decreased by 20 percent.  
23 The REA did not evaluate broad sensitive regions. However, in the sugar maple case study area  
24 (Kane Experimental Forest), recent deposition levels are associated with a BC:Al ratio below  
25 1.2, indicating between intermediate and low level of protection, which would indicate the  
26 potential for a greater than 20 percent reduction in growth. In the red spruce case study area  
27 (Hubbard Brook Experimental Forest), recent deposition levels are associated with a BC:Al ratio  
28 slightly above 1.2, indicating slightly better than an intermediate level of protection (**REA**  
29 **Section 4.3.5.1**).

30 Over the full range of sugar maple, 12 percent of evaluated forest plots exceeded the  
31 critical loads for a BC:Al ratio of 1.2, and 3 percent exceeded the critical load for a BC:Al ratio

1 of 0.6. However, there was large variability across states. In New Jersey, 67 percent of plots  
2 exceeded the critical load for a BC:Al ratio of 1.2, while in several states on the outskirts of the  
3 range for sugar maple (e.g. Arkansas, Illinois) no plots exceeded the critical load for a BC:Al  
4 ratio of 1.2. For red spruce, overall 5 percent of plots exceeded the critical load for a BC:Al ratio  
5 of 1.2, and 3 percent exceeded the critical load for a BC:Al ratio of 0.6. In the major red spruce  
6 producing states (Maine, New Hampshire, and Vermont), critical loads for a BC:Al ratio of 1.2  
7 were exceeded in 0.5, 38, and 6 percent of plots.

8 The ISA reported one study (McNulty, 1997) that estimated 15 percent of U.S. forest  
9 ecosystems exceeded the critical loads for acidity for N and S deposition by >250 eq/ha/year  
10 under current conditions (**ISA Section 4.2.1.3**). Staff concludes that this represents a significant  
11 portion of sensitive terrestrial ecosystems.

12 It can therefore be concluded that recent levels of NO<sub>x</sub> and SO<sub>x</sub> are associated with  
13 deposition that leads to BC:Al values below benchmark values that cause ecological harm in  
14 some sensitive terrestrial ecosystems. While effects are more widespread for sugar maple, there  
15 are locations with low to intermediate levels of protection from effects on both sugar maple and  
16 red spruce. While there are many other ecosystem services, including timber production, natural  
17 habitat provision, and regulation of water, climate, and erosion, potentially affected by  
18 reductions in BC:Al, linkages of BC:Al values to these additional ecosystem services is on the  
19 whole not well understood.

20 **4.5.2 To what extent does the current NO<sub>x</sub> secondary standard provide protection from**  
21 **adverse effects associated with deposition of atmospheric NO<sub>x</sub>, which results in nutrient**  
22 **enrichment effects in sensitive aquatic and terrestrial ecosystems?**

23 Nutrient enrichment effects are due to nitrogen loadings from both atmospheric and non-  
24 atmospheric sources. Evaluation of nutrient enrichment effects requires an understanding that  
25 nutrient inputs are essential to ecosystem health. The specific long term levels of nutrients in a  
26 system affect the types of species that occur over long periods of time. Short term additions of  
27 nutrients can affect species competition, and even small additions of nitrogen in areas that are  
28 traditionally nutrient poor can have significant impacts. In certain limited situations, additions of  
29 nitrogen can increase rates of growth, and these increases can have short term benefits in certain  
30 managed ecosystems. As noted earlier, this review of the standards is focused on unmanaged

1 ecosystems. As a result, in assessing adequacy of the current standards, we are focusing on the  
2 adverse effects of nutrient enrichment in unmanaged ecosystems. However, the following  
3 discussion provides a brief assessment of effects in managed ecosystems.

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

14       Information on the effects of changes in passive nitrogen deposition on forestlands and  
15 other terrestrial ecosystems is very limited. The multiplicity of factors affecting forests, including  
16 other potential stressors such as ozone, and limiting factors such as moisture and other nutrients,  
17 confound assessments of marginal changes in any one stressor or nutrient in forest ecosystems.  
18 The ISA notes that only a fraction of the deposited nitrogen is taken up by the forests, most of  
19 the nitrogen is retained in the soils (**ISA 3.3.2.1**). In addition, the ISA indicates that forest  
20 management practices can significantly affect the nitrogen cycling within a forest ecosystem, and  
21 as such, the response of managed forests to NO<sub>x</sub> deposition will be variable depending on the  
22 forest management practices employed in a given forest ecosystem (**ISA Annex C C.6.3**)  
23 Increases in the availability of nitrogen in N-limited forests via atmospheric deposition could  
24 increase forest production over large non-managed areas, but the evidence is mixed, with some  
25 studies showing increased production and other showing little effect on wood production (**ISA**  
26 **3.3.9**). Because leaching of nitrate can promote cation losses, which in some cases create nutrient  
27 imbalances, slower growth and lessened disease and freezing tolerances for forest trees, the net  
28 effect of increased N on forests in the U.S. is uncertain (**ISA 3.3.9**).

29       In managed agricultural ecosystems, nitrogen inputs from atmospheric NO<sub>x</sub> comprise a  
30 small fraction (less than 3 percent) of total nitrogen inputs, which include commercially applied  
31 fertilizers as well as applications of composted manure. And because of the temporal and spatial

1 variability in atmospheric deposition of NO<sub>x</sub>, it is unlikely that farmers would alter their  
2 fertilization decisions based on expected nitrogen inputs from NO<sub>x</sub>. And, in some locations,  
3 farmers need less nitrogen inputs due to production of excess nitrogen through livestock. In  
4 some locations, nitrogen production through livestock waste exceeds the absorptive capacity of  
5 the surrounding land, and as such, excess nitrogen from deposition of NO<sub>x</sub> in those locations  
6 reduces the capacity of the system to dispose of excess nitrogen, potentially increasing the costs  
7 of waste management from livestock operations (Letson and Gollehon, 1996). A USDA  
8 Economic Research Service report found that in 1997, 68 counties with high levels of confined  
9 livestock production had manure nitrogen levels that exceed the assimilative capacity of the  
10 entire county's crop and pasture land (Gollehon et al, 2001). In those locations, additional  
11 nitrogen inputs from NO<sub>x</sub> deposition will result in excess nitrogen, leading to nitrogen leaching  
12 and associated effects that adversely effect ecosystems.

13

#### 14 **4.5.3 Aquatic Nutrient Enrichment**

15 The REA case studies focused on coastal estuaries and revealed that while current  
16 ambient loadings of atmospheric NO<sub>x</sub> are contributing to the overall depositional loading of  
17 coastal estuaries, other non-atmospheric sources are contributing in far greater amounts in total,  
18 although atmospheric contributions are as large as some other individual source types. The  
19 ability of current data and models to characterize the incremental adverse impacts of nitrogen  
20 deposition is limited, both by the available ecological indicators, and by the inability to attribute  
21 specific effects to atmospheric sources of nitrogen. The REA case studies used as the ecological  
22 indicator for aquatic nutrient enrichment an index of eutrophication known as the Assessment of  
23 Estuarine Trophic Status Eutrophication Index (ASSETS EI). This index is a six level index  
24 characterizing overall eutrophication risk in a waterbody. This indicator is not sensitive to  
25 relatively large changes in nitrogen deposition. In addition, this type of indicator does not reflect  
26 the impact of nitrogen deposition in conjunction with other sources of nitrogen.

27 For example, if NO<sub>x</sub> deposition is contributing nine tenths of the nitrogen loading  
28 required to move a waterbody from an ASSETS EI category of "moderate" to a category of  
29 "poor", zeroing out NO<sub>x</sub> deposition will have no impact on the ASSETS EI value. However, if  
30 an area were to decide to put in place decreases in nitrogen loadings to move that waterbody

1 from “poor” to “moderate,” the area would have to reduce the full amount of the loadings  
2 through other sources if atmospheric deposition were not considered. Thus, the adverse impact  
3 of atmospheric nitrogen is in its contribution to the overall loading, and reductions in NO<sub>x</sub> will  
4 decrease the amount of reductions from other sources of nitrogen loadings that would be required  
5 to move from a lower ASSETS EI category to a higher category. NO<sub>x</sub> deposition can also be  
6 characterized as reducing the *risk* of a waterbody moving from a higher ASSETS EI category to  
7 a lower category, by reducing the vulnerability of that waterbody to increased loadings from  
8 non-atmospheric sources.

9 Based on the above considerations, staff preliminarily concludes that the ASSETS EI is  
10 not an appropriate ecological indicator for estuarine aquatic eutrophication. Staff further  
11 concludes that additional analysis is required to develop an appropriate indicator for determining  
12 the appropriate levels of protection from N nutrient enrichment effects in estuaries related to  
13 deposition of NO<sub>x</sub>. As a result, staff is unable to make a determination as to the adequacy of the  
14 existing secondary NO<sub>x</sub> standard in protecting public welfare from N nutrient enrichment effects  
15 in estuarine aquatic ecosystems.

16 Additionally, nitrogen deposition can alter species composition and cause eutrophication  
17 in freshwater systems. In the Rocky Mountains, for example, deposition loads of 1.5 to 2 kg/ha-  
18 yr which are well within current ambient levels are known to cause changes in species  
19 composition in diatom communities indicating impaired water quality (**ISA Section 3.3.5.3**). It  
20 then seems apparent then that the existing secondary standard for NO<sub>x</sub> does not protect such  
21 ecosystems and their resulting services from impairment.

#### 22 **4.5.4 Terrestrial Nutrient Enrichment**

23 The scientific literature has many examples of the deleterious effects caused by excessive  
24 nitrogen loadings to terrestrial systems. Several studies have set benchmark values for levels of  
25 N deposition at which scientifically adverse effects are known to occur. These benchmarks are  
26 discussed more thoroughly in Chapter 5 of the REA. Large areas of the country appear to be  
27 experiencing deposition above these benchmarks for example, Fenn et al. (2008) found that at  
28 3.1 kg N/ha-yr, the community of lichens begins to change from acidophytic to tolerant species;  
29 at 5.2 kg N/ha-yr, the typical dominance by acidophytic species no longer occurs; and at 10.2 kg  
30 N/ha-yr, acidophytic lichens are totally lost from the community. Additional studies in the

1 Colorado Front Range of the Rocky Mountain National Park support these findings and are  
2 summarized in Chapter 6.0 of the *Risk and Exposure Assessment*. These three values (3.1, 5.2,  
3 and 10.2 kg/ha-yr) are one set of ecologically meaningful benchmarks for the mixed conifer  
4 forest (MCF) of the pacific coast regions. Nearly all of the known sensitive communities receive  
5 total nitrogen deposition levels above the 3.1 N kg/ha-yr ecological benchmark according to  
6 the 12 km, 2002 CMAQ/NADP data, with the exception of the easternmost Sierra Nevadas.  
7 MCFs in the southern portion of the Sierra Nevada forests and nearly all MCF communities in  
8 the San Bernardino forests receive total nitrogen deposition levels above the 5.2 N kg/ha-yr  
9 ecological benchmark.

10 Coastal Sage Scrub communities (CSS) are also known to be sensitive to community  
11 shifts caused by excess nitrogen loadings. Wood et al. (2006) investigated the amount of  
12 nitrogen utilized by healthy and degraded CSS systems. In healthy stands, the authors estimated  
13 that 3.3 kg N/ha-yr was used for CSS plant growth (Wood et al., 2006). It is assumed that 3.3 kg  
14 N/ha-yr is near the point where nitrogen is no longer limiting in the CSS community. Therefore,  
15 this amount can be considered an ecological benchmark for the CSS community. The majority of  
16 the known CSS range is currently receiving deposition in excess of this benchmark. Thus, staff  
17 concludes that recent conditions where NO<sub>x</sub> ambient concentrations are at or below the current  
18 NO<sub>x</sub> secondary standards are not adequate to protect against anticipated adverse impacts from N  
19 nutrient enrichment in sensitive ecosystems.

#### 20 21 **4.6 To What Extent Do The Current NO<sub>x</sub> And/Or SO<sub>x</sub> Secondary Standards Provide** 22 **Protection From Other Ecological Effects (Eg. Mercury Methylation) Associated** 23 **With The Deposition Of Atmospheric NO<sub>x</sub>, And/Or SO<sub>x</sub>?** 24

25 It is stated in the ISA (**ISA Sections 3.4.1 and 4.5**) that mercury is a highly neurotoxic  
26 contaminant that enters the food web as a methylated compound, methylmercury. Mercury is  
27 principally methylated by sulfur-reducing bacteria and can be taken up by microorganisms,  
28 zooplankton and macroinvertebrates. The contaminant is concentrated in higher trophic levels,  
29 including fish eaten by humans. Experimental evidence has established that only inconsequential  
30 amounts of methylmercury can be produced in the absence of sulfate. Once methylmercury is  
31 present, other variables influence how much accumulates in fish, but elevated mercury levels in

1 fish can only occur where substantial amounts of methylmercury are present. Current evidence  
2 indicates that in watersheds where mercury is present, increased SO<sub>x</sub> deposition very likely  
3 results in additional production of methylmercury which leads to greater accumulation of MeHg  
4 concentrations in fish (Munthe et al, 2007; Drevnick et al., 2007).

5         The production of meaningful amounts of methylmercury (MeHg) requires the presence  
6 of SO<sub>4</sub><sup>2-</sup> and mercury, and where mercury is present, increased availability of SO<sub>4</sub><sup>2-</sup> results in  
7 increased production of MeHg. There is increasing evidence on the relationship between sulfur  
8 deposition and increased methylation of mercury in aquatic environments; this effect occurs only  
9 where other factors are present at levels within a range to allow methylation. The production of  
10 methylmercury requires the presence of sulfate and mercury, but the amount of methylmercury  
11 produced varies with oxygen content, temperature, pH, and supply of labile organic carbon (**ISA**  
12 **Section 3.4**). In watersheds where changes in sulfate deposition did not produce an effect, one or  
13 several of those interacting factors were not in the range required for meaningful methylation to  
14 occur (**ISA Section 3.4**). Watersheds with conditions known to be conducive to mercury  
15 methylation can be found in the northeastern United States and southeastern Canada. The  
16 relationship between sulfur and methylmercury production is addressed qualitatively in Chapter  
17 6 of the Risk and Exposure Assessment.

18         With respect to sulfur deposition and mercury methylation, the final ISA determined: *The*  
19 *evidence is sufficient to infer a causal relationship between sulfur deposition and increased*  
20 *mercury methylation in wetlands and aquatic environments.* However, staff did not conduct a  
21 quantitative assessment of the risks associated with increased mercury methylation under current  
22 conditions. As such, staff are unable to make a determination as to the adequacy of the existing  
23 SO<sub>2</sub> standards in protecting against welfare effects associated with increased mercury  
24 methylation.

25

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## 5 OPTIONS FOR ELEMENTS OF THE STANDARD

The elements of the standard include the ambient air indicator, the form, the averaging time and the level. The “indicator” of a standard defines the chemical species or mixture of criteria air pollutants that is to be measured in determining whether an area attains the standard. The “form” of a standard defines the air quality statistic that is to be compared to the level of the standard in determining whether an area attains the standard. The “averaging time” defines the period of time over which the air quality indicator is averaged, e.g. annual average. The “level” is the specific quantity to which the air quality statistic will be compared.

Historically, EPA has established NAAQS so that the locally-monitored ambient concentration of an air pollutant indicator is compared against a specified numerical level of atmospheric concentration, using a specified statistical form and averaging time. For example, the current secondary standard for oxides of nitrogen uses ambient concentrations of  $\text{NO}_2$  as the indicator. Attainment is determined by comparing the annual arithmetic mean of the measured maximum daily 1-hour  $\text{NO}_2$  concentrations, for a calendar year, against the level of 0.053 ppm. As discussed in Chapters 4, a standard using this kind of approach for defining indicator, form, averaging time, and level is not the most appropriate way to protect sensitive ecosystems from effects associated with ambient concentrations of  $\text{NO}_x$  and  $\text{SO}_x$ . This is because the ecological effects of  $\text{NO}_x$  and  $\text{SO}_x$  are a result of deposition of these air pollutants. The inherently complex and variable linkages between ambient concentrations of  $\text{NO}_x$  and  $\text{SO}_x$ , their deposited forms of nitrogen and sulfur, and the ecological responses that are associated with public welfare effects call for consideration of a more complex and ecologically relevant design of the standard that reflects these linkages. In this chapter, we present a set of potential options for defining the elements of the  $\text{NO}_x$  and  $\text{SO}_x$  secondary standard including indicator, form, averaging time, and certain aspects of the level, with additional discussion of the options for specifying a range of levels is discussed in Chapter 9.

After review of the ISA and REA, CASAC concluded that aquatic acidification should be the focus for developing a multi-pollutant standard, based on the quantity and quality of available data. CASAC also recommended that, in addition to aquatic acidification, the EPA

1 should consider multiple ecological indicators and made the following statement in their letter to  
2 the EPA on August 28, 2009:

3  
4 “...the Panel finds the information in the current REA sufficient to inform setting  
5 separate standards for terrestrial acidification, eutrophication of western alpine lakes and  
6 terrestrial nutrient enrichment. However, the Panel believes that setting a standard for  
7 coastal nutrient enrichment would be difficult because of the substantial inputs of non-  
8 atmospheric sources of N to these systems.”  
9

10 As a result of our assessment of the science, and reflecting the comments of CASAC, this policy  
11 assessment is focused on developing a standard specifically designed to protect against the  
12 effects of aquatic acidification in sensitive ecosystems, while recognizing that such a standard  
13 may also provide co-protection against effects of terrestrial acidification, eutrophication of high  
14 elevation western lakes and terrestrial nutrient enrichment. Co-protection against these effects is  
15 discussed in Chapter 6.

16 Our development of options for the standards recognizes the need for nationally  
17 applicable standard for protection against adverse effects to public welfare, while recognizing the  
18 complex and heterogeneous interactions between atmospheric concentrations of NO<sub>y</sub> and SO<sub>x</sub>,  
19 deposition from of NO<sub>y</sub> and SO<sub>x</sub>, and ecological response. Our approach also recognizes that  
20 while a standard is national in scope and coverage, the effects to public welfare from aquatic  
21 acidification may not occur to the same extent in all locations in the U.S. – in fact, protection  
22 may vary among locations according to sensitivity. As noted in Chapters 2, 3, and 4, many  
23 locations in the U.S. are naturally protected against acid deposition due to underlying geological  
24 conditions. Likewise, some locations in the U.S., including lands managed for commercial  
25 agriculture and forestry, are not likely to be negatively impacted by current levels of nitrogen and  
26 sulfur deposition. As a result, our design for the standards is intended to protect sensitive  
27 ecosystems and the services provided by those sensitive ecosystems.

28 In this chapter we present our reasoning for suggesting a standard that employs (1) NO<sub>y</sub>  
29 and SO<sub>x</sub> as the atmospheric indicators, (2) a multi-year averaging time, (3) a form of a secondary  
30 standard that takes into account variable factors, such as atmospheric and ecosystem conditions  
31 that modify the amounts of deposited NO<sub>x</sub> and SO<sub>x</sub>, and the associated effects of deposited N  
32 and S on ecosystems, and (4) a target ANC level and target percentages of water bodies to  
33 protect to the target ANC level. Our goal in developing the form of the standard is to create an

1 index, directly expressed in terms of atmospheric concentrations of NO<sub>y</sub> and SO<sub>x</sub>, which can be  
2 applied across the nation to convey the allowable levels of ambient NO<sub>y</sub> and SO<sub>x</sub>, based on  
3 various factors such as the sensitivity of an area and the desired degree of protection from  
4 acidification caused by atmospheric deposition. This chapter is structured around questions  
5 related to the various elements of a standard. The chapter begins in section 5.1 with a discussion  
6 of atmospheric indicators. In Section 5.2 the averaging times for the atmospheric indicators is  
7 presented. In Section 5.3 a suggested ecologically relevant form of the standard is presented.  
8 In Section 5.4 the issues regarding the spatial area over which a standard might be evaluated are  
9 discussed, along with related issues regarding spatial averaging within areas. Section 5.5  
10 provides a discussion of specific target ANC levels. Section 5.6 discusses the selection of target  
11 percentages of water bodies to protect to this ANC level. As noted previously, additional  
12 discussion related to options for ranges of the level of the standard are discussed in Chapter 9.

13

14 **5.1 What atmospheric indicators of oxidized nitrogen and sulfur are appropriate for**  
15 **use in a secondary NAAQS that provides protection for public welfare from**  
16 **exposure related to deposition of NO<sub>x</sub> and SO<sub>x</sub>?**

17

18 Staff concludes that indicators other than NO<sub>2</sub> and SO<sub>2</sub> should be considered as the  
19 appropriate pollutant indicators for protection against the acidification effects associated with  
20 deposition of NO<sub>x</sub> and SO<sub>x</sub>. **This conclusion is based on the recognition that all forms of**  
21 **oxidized nitrogen and sulfur in the atmosphere contribute to deposition and resulting**  
22 **acidification, and as such NO<sub>2</sub> and SO<sub>2</sub> are incomplete indicators. Furthermore, staff**  
23 **concludes that NO<sub>y</sub> (total oxidized nitrogen) should be considered as an appropriate indicator**  
24 **for oxides of nitrogen. NO<sub>y</sub> is defined as NO<sub>x</sub> (NO and NO<sub>2</sub>) and all oxidized NO<sub>x</sub> products:**  
25 **including NO, NO<sub>2</sub>, and all other oxidized N-containing compounds transformed from**  
26 **NO and NO<sub>2</sub> (Finlayson-Pitts and Pitts, 2000). As described in Chapter 4, this set of**  
27 **compounds includes NO<sub>2</sub> + NO + HNO<sub>3</sub> + PAN + 2N<sub>2</sub>O<sub>5</sub> + HONO + NO<sub>3</sub> + organic nitrates +**  
28 **particulate NO<sub>3</sub>. SO<sub>x</sub> includes sulfur monoxide (SO), sulfur dioxide, sulfur trioxide (SO<sub>3</sub>),**  
29 **and disulfur monoxide (S<sub>2</sub>O), and particulate-phase S compounds that result from gas-**  
30 **phase sulfur oxides interacting with particles. Staff concludes that SO<sub>2</sub> + SO<sub>4</sub> should be**  
31 **considered as an appropriate indicator for oxides of sulfur.**

1 In principle, measured NO<sub>y</sub> based on catalytic conversion of all oxidized species to NO,  
2 followed by chemiluminescence NO detection is consistent with this definition. We recognize  
3 the caveats associated with instrument conversion efficiency and possible inlet losses which are  
4 discussed in Chapter 8. The development of the function that converts atmospheric  
5 concentrations of NO<sub>y</sub> and SO<sub>x</sub> to N and S deposition incorporates NO<sub>y</sub> estimates based on the  
6 Community Multi-scale Air Quality (CMAQ) model (EPA, 1999). CMAQ treats the dominant  
7 NO<sub>y</sub> species (NO, NO<sub>2</sub>, nitric acid, PAN and particulate NO<sub>3</sub>) as explicit species while the  
8 minor contributing non-PAN organic nitrogen compounds are aggregated. From a  
9 measurement and modeling perspective we only consider the sum of SO<sub>2</sub> and particulate SO<sub>4</sub> as  
10 the indicator for sulfur. The sum of SO<sub>2</sub> and SO<sub>4</sub> constitute virtually all of the ambient air  
11 sulfur budget and are measured routinely in monitoring networks. In addition to accounting for  
12 virtually the entire oxidized sulfur budget, SO<sub>2</sub> and particulate SO<sub>4</sub> are routinely measured in  
13 ambient air monitoring networks, although only CASTNET filter packs capture the entire  
14 particle size range. The CMAQ treatment of SO<sub>x</sub> is the simple addition of both species which  
15 are treated explicitly in the model formulation. All particle size fractions are included in the  
16 CMAQ SO<sub>x</sub> estimates. Consistent with units and the charge balance relationships applied in  
17 ecosystem acidification models, only mass as sulfur or nitrogen is considered when aggregating  
18 the species constituting NO<sub>y</sub> or SO<sub>x</sub>.

19  
20 **5.2 What is the appropriate averaging time for the air quality indicators NO<sub>y</sub> and SO<sub>x</sub>**  
21 **to provide protection of public welfare from adverse effects from aquatic**  
22 **acidification?**  
23

24 Based on the review of the scientific evidence, welfare effects associated with  
25 acidification result from event based to annual cumulative deposition of N and S. Annual  
26 cumulative deposition of N and S reflects the chronic acid base balance of the surface water as  
27 indicated by the ANC level (measured as annual ANC). Also, critical loads for acidity are in  
28 terms of annual cumulative deposition of N and S. Aquatic acidification can occur over both  
29 long- and short-term timescales. Short-term (i.e., hours or days) episodic changes in water  
30 chemistry, often due to changes in the hydrologic flow paths (Chen et al. 1984), can have  
31 significant biological effects. Short-term change in chemistry is termed “episodic acidification.”

1 Some streams may have chronic or base flow chemistry that is suitable for aquatic biota, but may  
2 be subject to occasional acidic episodes with lethal consequences.

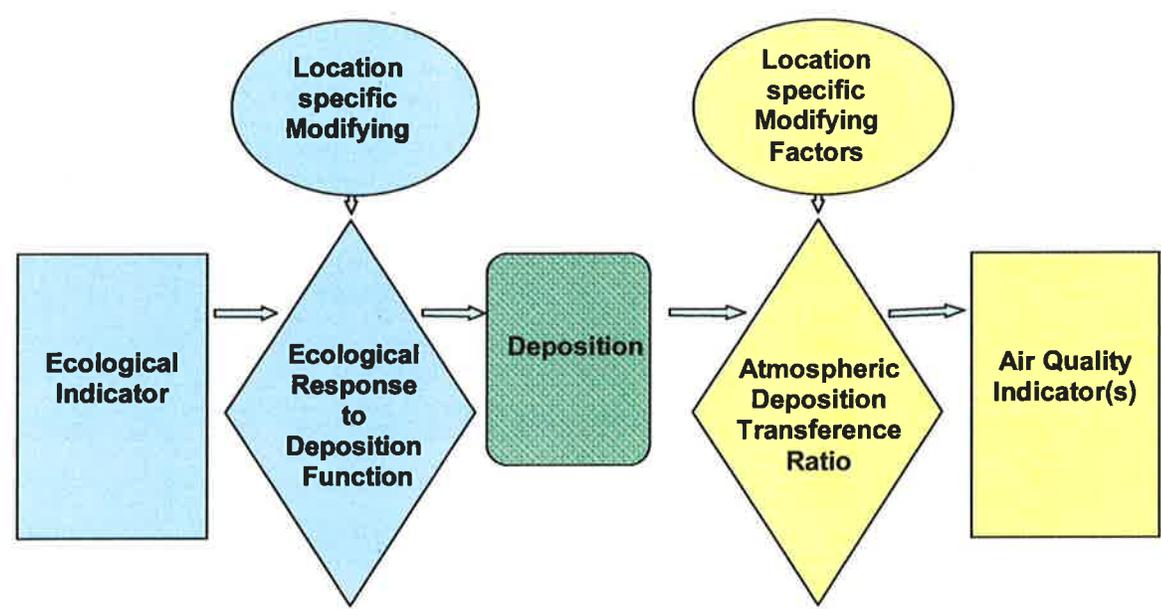
3 Episodic declines in pH and ANC are nearly ubiquitous in drainage waters throughout the  
4 eastern United States. Episodic acidification can result from several mechanisms related to  
5 changes in hydrologic flow paths. For example, snow can store N deposited throughout the  
6 winter, snowmelt releases this stored N in a pulse that leads to episodic acidification in the  
7 absence of increased deposition during the actual episodic acidification event. However, inputs  
8 of nitrogen and sulfur from snowpack and atmospheric deposition largely cycle through soil. As  
9 a result short-term direct deposition inputs are not important in episodic acidification. As noted  
10 in Chapter 3 of the ISA, protection against episodic acidity events can be achieved by  
11 establishing a higher chronic ANC level (See 5.3.2.1). Protection against a low chronic ANC  
12 level is provided by reducing overall annual average deposition levels for nitrogen and sulfur.  
13 This supports the conclusion that long term NO<sub>y</sub> and SO<sub>x</sub> concentrations are appropriate to  
14 provide protection against low chronic ANC levels, which protects against both long term  
15 acidification and acute acidic episodes.

16 Staff suggests a 3 to 5 year averaging time of the cumulative annual average for the  
17 air quality indicators NO<sub>y</sub> and SO<sub>x</sub>, to account for interannual variations.

18  
19 **5.3 What form(s) of the standard are most appropriate to provide protection of**  
20 **sensitive ecosystems from the effects of acidifying deposition related to ambient NO<sub>x</sub>**  
21 **and SO<sub>x</sub> concentrations?**  
22

23 Based on the evidence of the aquatic acidification effects caused by NO<sub>y</sub> and SO<sub>x</sub>, staff  
24 concludes that it is appropriate to consider changes to the form of the existing NO<sub>x</sub> and SO<sub>x</sub>  
25 secondary standards to provide protection to ecosystems. EPA staff has developed a conceptual  
26 design for the form of the standard that includes four main components: atmospheric and  
27 ecological indicators, deposition metrics, functions that relate indicators to deposition metrics  
28 and factors that modify the functions. These components of the design are illustrated in Fig 5.1.  
29 The rectangles represent indicators. Ecological indicators are chemical or biological components  
30 of the ecosystem that can be linked to N and S deposition based on scientific evidence. Air  
31 quality indicators are the chemical species of the criteria air pollutants that best represent the  
32 atmospheric pollutants that cause ecological harm in the criteria pollutant categories of oxides of

1 nitrogen and oxides of sulfur (selection of air quality indicators was discussed in Section 5.1).  
 2 Diamonds indicate functions in which two variables are related. The ecological effect function is  
 3 the relationship between the ecological indicator and deposition. The atmospheric deposition  
 4 transference ratio is the relationship between deposition and the atmospheric concentration of an  
 5 air quality indicator. The ovals represent factors which will modify the functions. Modifying  
 6 factors can vary across the landscape, such as soil depth, catchment size, etc. The spatial  
 7 heterogeneity of modifying factors can be challenging to characterize, and therefore in some  
 8 cases we present multiple options for how to incorporate them into the design.

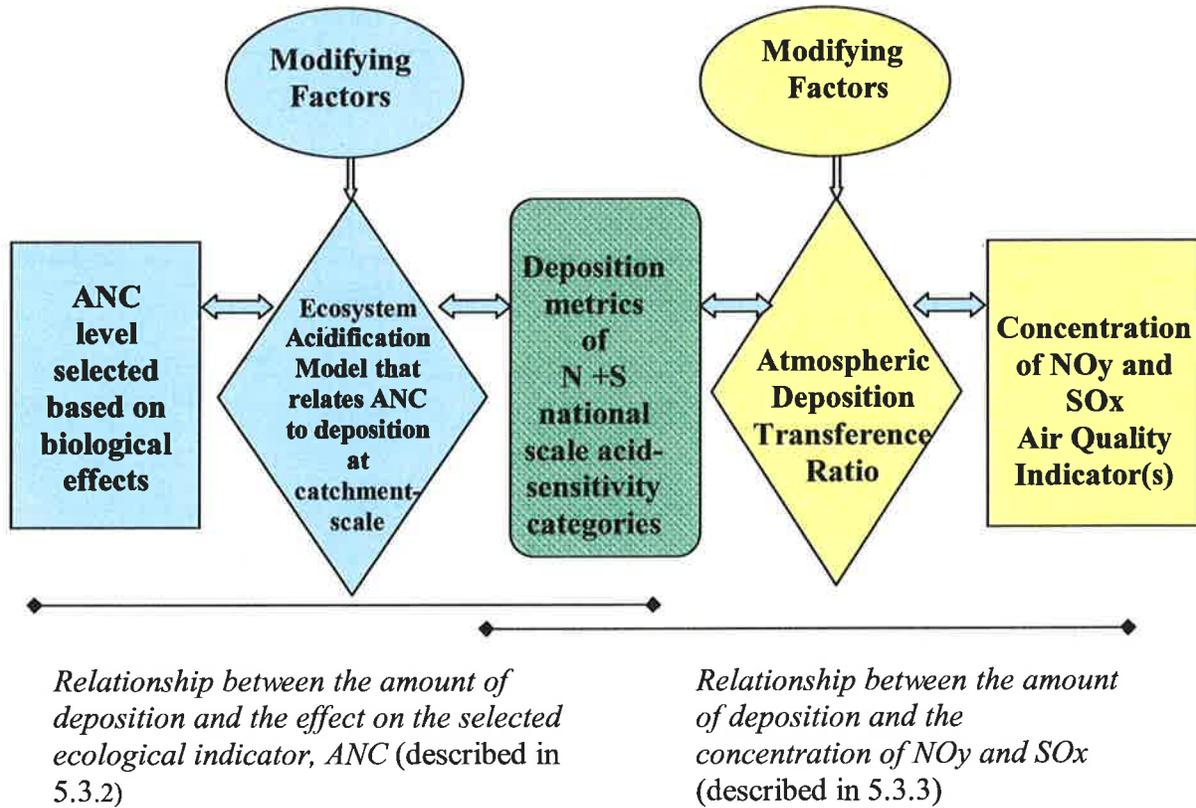


11  
 12  
 13 **Figure 5-1 Conceptual design of the form of NO<sub>x</sub> and SO<sub>x</sub> secondary standard**

14  
 15 **5.3.1 Conceptual Design of the Form: General Overview**

16 A summary of the design of the form based on aquatic acidification is given here to help  
 17 provide context and support for the more detailed discussions that follow. The conceptual design  
 18 for a form that is specifically based on aquatic acidification effects is illustrated by Fig 5.2.  
 19 Starting on the left side of the figure, ANC is a chemical indicator of sensitivity to aquatic  
 20 acidification that is tied to the probability of biological harm that may occur to the system from  
 21 aquatic acidification. ANC is often considered the best chemical indicator of sensitivity to  
 22 aquatic acidification (Section 5.3.2.1). Ambient NO<sub>y</sub> and SO<sub>x</sub> add to the total deposition of N  
 23 and S that lead to aquatic acidification. NH<sub>x</sub> is a component comprising a varying fraction of the

1 total N deposition. The load of N and S deposition that causes a specific level of ANC will vary  
 2 depending on the characteristics of the receiving ecosystem, but can be quantified at the  
 3 catchment scale with acid-base balance acidification models. The acidification models that  
 4 characterize the relationship between ANC and N and S deposition that are most appropriate to  
 5 inform the NAAQS are discussed in Sections 5.3.2.  
 6



7 **Figure 5-2. Conceptual design of the form of the standard based on**  
 8 **aquatic acidification.**

9 Catchment scale modeling is necessary to understand the sensitivity of individual  
 10 waterbodies to acid deposition. Secondary NAAQS, however, are intended to be requisite to  
 11 protect public welfare from known or anticipated adverse effects. One aspect of adversity to  
 12 public welfare is a consideration of the requisite amount of protection for sensitive populations  
 13 or groups of catchments. In so doing, the Administrator seeks to establish standards that are  
 14 neither more nor less stringent than necessary for this purpose. In order to establish such a  
 15 standard for the entire country, it is necessary to look across variability both in effect and

1 exposure. In addition, there are uncertainties in our estimates of risk and exposure for individual  
2 waterbodies. Therefore, we propose to examine various alternative spatial groupings of sensitive  
3 populations of waterbodies to inform the development of requisite protection. EPA staff  
4 proposes to categorize the landscape nationally, such that within a category there are generally  
5 similar acid-sensitivity characteristics. Each national acid-sensitivity category is represented by  
6 a population of catchments for which critical loads at a specified ANC limit are calculated.

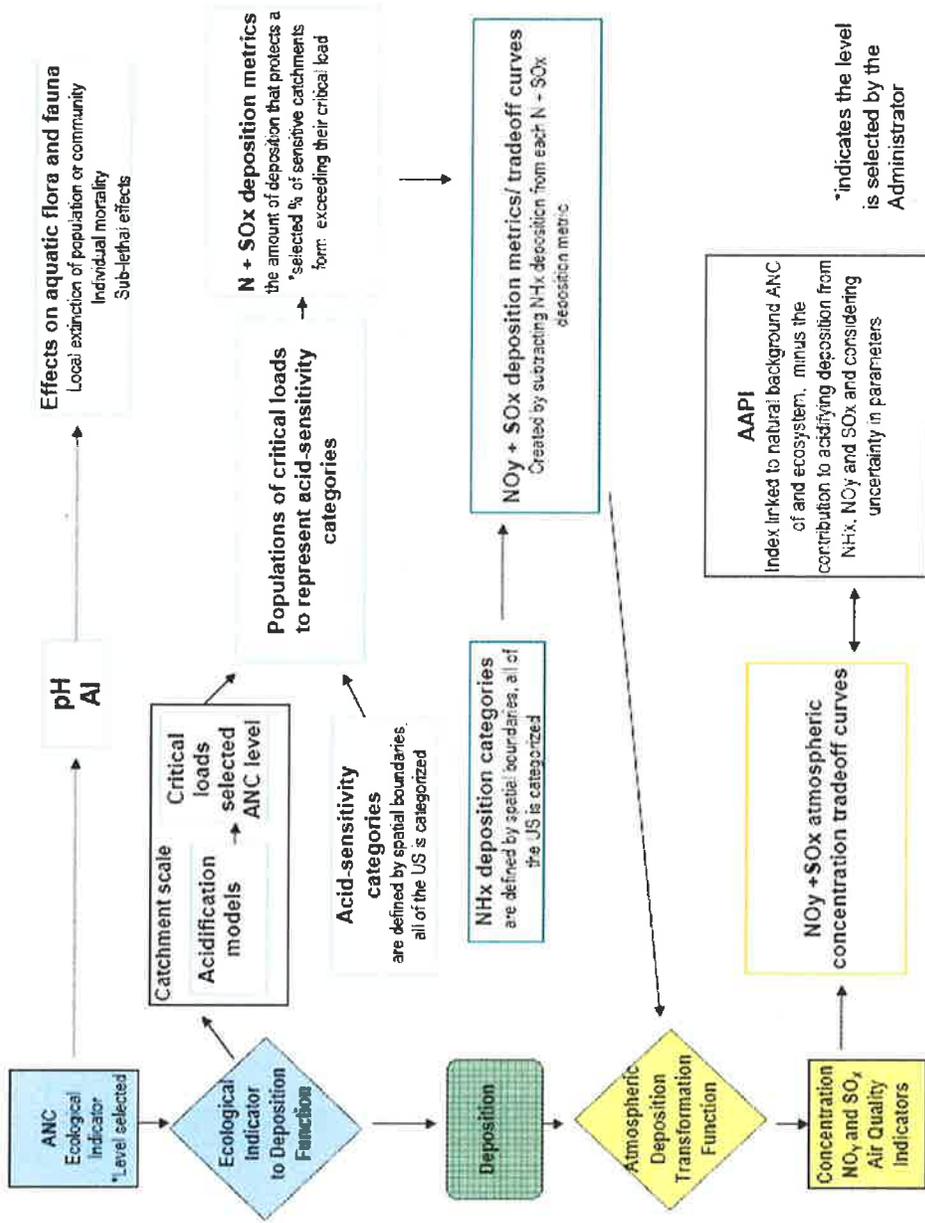
7 In our proposed approach, for each acid-sensitive category of the landscape the  
8 distribution of critical loads from a population of catchments would be evaluated to develop a  
9 deposition metric. The deposition metric would be defined as the amount of deposition that  
10 reasonably assures a specified percentage of individual catchments in a population would  
11 achieve a target ANC level.

12 The level of the ANC target is tied to the described degree of protection for aquatic biota.  
13 It will be selected as one element of the set of information that informs the Administrator's  
14 selection of the allowable level(s) of ambient NO<sub>y</sub> and SO<sub>x</sub> under the secondary standard.  
15 Options for target ANC levels are discussed in Section 5.5. Taken together, the secondary  
16 standard would be set with a goal of specifying allowable ambient levels of NO<sub>y</sub> and SO<sub>x</sub> such  
17 that there is a desired degree of confidence that a targeted percentage of aquatic ecosystems  
18 would achieve a specified ANC level within each acid-sensitivity category (see Chapter 9).

19 Finally, the deposition of reduced N should be taken into account because reduced forms  
20 of N are often quickly converted to NO<sub>3</sub><sup>-</sup> by the processes of nitrification. For this reason, the  
21 amount of reduced N needs to be accounted for in the standard because some fraction of the acid  
22 buffering capacity of an ecosystem is used up by reduced nitrogen, leaving less protection  
23 against deposition from NO<sub>y</sub> and SO<sub>x</sub>. Spatial characterization of reduced N is discussed in  
24 Section 5.3.2. The deposition tradeoff curves are then multiplied by the deposition to  
25 concentration transference ratio to calculate the atmospheric concentration tradeoff curves.  
26 Development of these functions is discussed in Section 5.3.3. Thus far we have summarized our  
27 conceptual design by walking through the components of the standard shown in Fig 5.2 from the  
28 left to the right, and a more detailed illustration of these steps is given by Fig 5.3.

29 The bidirectional arrows shown in Fig 5.2 emphasize that the order in which one  
30 considers the links between ANC and deposition resulting from of NO<sub>y</sub> and SO<sub>x</sub> is conceptually  
31 important. Moreover, different questions may be answered by working through Fig 5.2 right to

1 left versus the left to right. For example, if the amount of N and S deposited to a given  
2 catchment is known, the level of ANC that would result may be calculated. The calculated ANC  
3 could then be compared to a benchmark value of ANC. Working through Fig 5.2 from the right  
4 to the left is the basis for the Atmospheric Acidification Protection Index (AAPI, Fig 5.3 and  
5 Section 5.3.4). The AAPI is essentially a function that determines the allowable levels of  
6 ambient NO<sub>y</sub> and SO<sub>x</sub> based on the target ANC limit, given uncertainties in the parameters used  
7 to calculate an ANC equivalent at the national scale, and weighing other factors such as time to  
8 recovery for ecosystems, based on populations of catchments that represent acid-sensitive areas  
9 in the U.S. The AAPI is designed to be a more ecologically relevant form of the standard  
10 relative to the current form. The intent of the AAPI is to weight atmospheric concentrations of  
11 NO<sub>x</sub> and SO<sub>x</sub> by their propensity to contribute to acidification through deposition, given the  
12 fundamental acidifying potential of each pollutant, and the ecological factors that govern acid  
13 sensitivity in different ecosystems, as well as the contribution of reduced nitrogen. Thus the  
14 AAPI is more relevant to protecting ecosystems from acidifying deposition compared to simple  
15 ambient concentration forms which do not reflect factors that affect acidifying potential.



**Figure 5-3 Detailed conceptual design of the form of the standard based on aquatic acidification.**

1  
2  
3

1 Staff notes two important concepts illustrated by the O<sub>3</sub> and PM<sub>10</sub> NAAQS that  
2 lend support to using this index as the form of a NAAQS. First, in recent reviews  
3 of the secondary ozone standards, EPA has considered use of a form of the  
4 standard that reflects ecologically relevant exposures, by using a cumulative index  
5 which weights exposures at higher concentrations greater than those at lower  
6 concentrations based on scientific literature demonstrating the cumulative nature  
7 of O<sub>3</sub>-induced plant effects and the need to give greater weight to higher  
8 concentrations (EPA, 2007; See 75 FR 2938, 2999 January 19, 2010). Staff also  
9 notes that PM<sub>10</sub> is the indicator for the coarse PM NAAQS standard (PM<sub>10</sub> =  
10 PM<sub>2.5</sub> + PM<sub>10-2.5</sub>). Although the standard has a single level (150 μg/m<sup>3</sup>), the actual  
11 amount of coarse PM that is allowed varies depending on how much fine PM  
12 (PM<sub>2.5</sub>) is present. By its nature, the PM<sub>10</sub> NAAQS provides the appropriate level  
13 of protection from exposure to coarse PM across locations using a related index of  
14 PM (PM<sub>10</sub>) as the indicator, while allowing the, level of coarse PM to vary across  
15 locations. The proposed form for the NO<sub>x</sub> and SO<sub>x</sub> standard builds on the  
16 concept of using a related index (the AAPI) to provide a homogeneous level of  
17 protection across the nation, while allowing ambient air concentrations of NO<sub>x</sub>  
18 and SO<sub>x</sub> to vary based on ecosystem sensitivity and other relevant factors.

### 20 **5.3.2 Conceptual design of the form: Linking the ecosystem indicator to deposition**

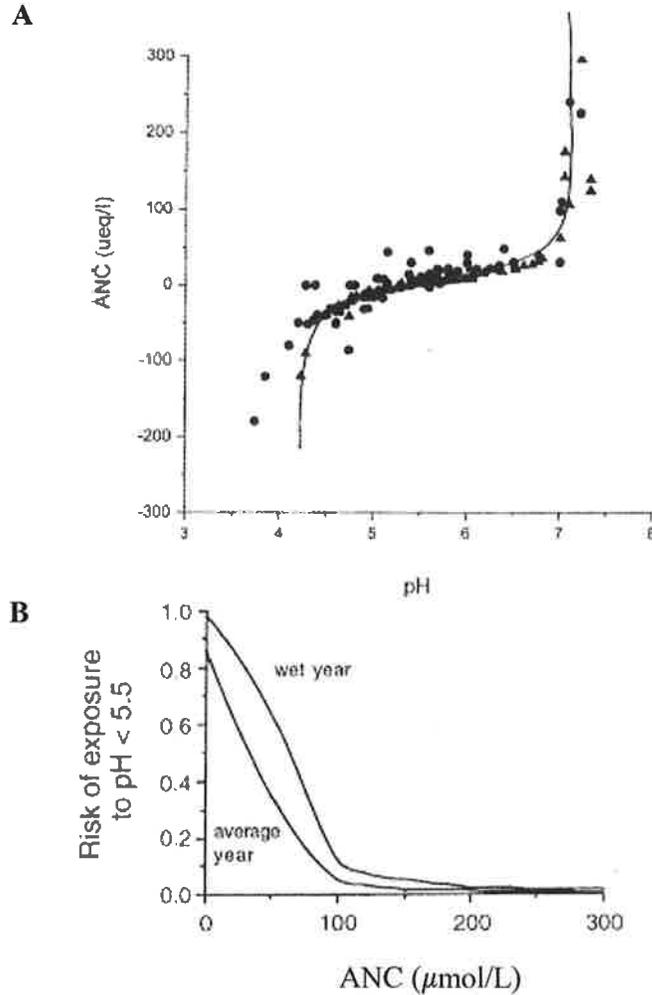
21 The relationship between the ecosystem indicator, acidification models, and  
22 deposition metric components of the form are discussed in this section. The relationship  
23 between deposition metric and atmospheric concentrations is discussed in Section 5.3.3.  
24 and the derivation of the AAPI equation is presented in Section 5.3.4.

#### 26 **5.3.2.1 Ecological Indicator:** *Does the available information provide support to use* 27 *ANC as the ecological indicator in the conceptual design of the NO<sub>y</sub> and SO<sub>x</sub> standard* 28 *based on aquatic acidification?*

30 Ecological indicators of acidification in aquatic ecosystems can be chemical or  
31 biological components of the ecosystem that are altered by the acidifying effects of N and  
32 S deposition. A desirable ecological indicator for aquatic acidification is one that is

1 measurable or estimable, linked causally to deposition of N and S, and linked causally,  
2 either directly or indirectly to ecological effects known or anticipated to adversely affect  
3 public welfare.

4 As summarized in Chapter 2, atmospheric deposition of NO<sub>y</sub> and SO<sub>x</sub> causes  
5 aquatic acidification through the input of strong acid anions (e.g. NO<sub>3</sub><sup>-</sup> and SO<sub>4</sub><sup>2-</sup>) or  
6 chemical forms that are transformed by the ecosystem into strong acid ions. The anions  
7 are deposited either directly to the aquatic ecosystem, or indirectly via drainage from  
8 terrestrial ecosystems. In other words, when these anions are mobilized in the terrestrial  
9 soil, they can leach into adjacent waterbodies. Aquatic acidification is indicated by  
10 changes in the surface water chemistry of ecosystems. In turn, the alteration of surface  
11 water chemistry has been linked to negative effects on the biotic integrity of freshwater  
12 ecosystems. There are a suite of chemical indicators that could be used to assess the  
13 effects of acidifying deposition on lake or stream acid-base chemistry. These indicators  
14 include acid neutralizing capacity (ANC), surface water pH and concentrations of SO<sub>4</sub><sup>2-</sup>,  
15 NO<sub>3</sub><sup>-</sup>, Al, and Ca<sup>2+</sup>; the sum of base cations; and the base cation surplus. ANC is the  
16 most widely used chemical indicator of acid sensitivity and has been found in various  
17 studies to be the best single indicator of the likelihood of biological response and health  
18 of aquatic communities in acid-sensitive systems (Lien et al., 1992; Sullivan et al., 2006).  
19 The utility of the ANC criterion lies in the association between ANC and the surface  
20 water constituents that directly contribute to or ameliorate acidity-related stress, in  
21 particular pH, Ca<sup>2+</sup>, and Al. For example, there is an extensive literature documenting  
22 threshold pH levels for various aquatic species mortality and health. ANC and pH are  
23 directly related under equilibrium conditions (Fig 5.4A). In the field there is a  
24 relationship between decreasing ANC and increasing risk to exposure of a threshold pH  
25 level (Fig 5.4B). ANC is also used because it integrates overall acid status (**ISA 3.2.3**  
26 **and REA 5.2.1**) and the acid-related stress for biota that occupies the water that can be  
27 directly related to biological impairment, including the number of fish species (**ISA**  
28 **3.2.3**).



ANC

1

2 **Figure 5-4.** A. The relationship between pH and ANC under equilibrium conditions  
 3 with mineral phase gibbsite. Triangles indicate calculated values while  
 4 circles indicate measurements (Bi and Liu 2001). B. The relationship  
 5 between precipitation (wet and average year), ANC and the risk to  
 6 exposure of a pH below 5.5 (Gerritsen et al. 1996).

7

8 EPA staff thus concludes that the available information provides support for the  
 9 use of ecological indicators to characterize the responses of aquatic ecosystems to  
 10 acidification effects associated with N and S deposition, and that ANC is the most  
 11 supportable indicator.

12

1 **5.3.2.2 Relating the ecological indicator to atmospheric deposition: Does a**  
2 *quantified relationship exist between the ecological indicator and atmospheric*  
3 *deposition of nitrogen and sulfur?*  
4

5 There is evidence to support a quantified relationship between deposition of N  
6 and S, and ANC. This relationship was analyzed in the REA to determine current risk for  
7 two case study areas, the Adirondack and Shenandoah Mountains, with three techniques  
8 (1) a time series analysis to evaluate long-term trends using MAGIC modeling to  
9 reconstruct past trends, (2) a time series analysis using monitoring data to evaluate recent  
10 observed trends and (3) a critical load approach using water chemistry data from the  
11 Temporally Integrated Monitoring of Ecosystems (TIME) program and Long-term  
12 Monitoring (LTM) to calculate critical loads with the SSWC and FAB aquatic  
13 acidification models. A technical summary of the methods used in the REA are provided  
14 in Appendix A.

15 **Modeled long-term trends over time**

16 In the REA analysis, long-term trends in surface water nitrate, sulfate and ANC  
17 were modeled using Model of Acidification of Groundwater in Catchment (MAGIC) for  
18 the two case study areas. These data were used to compare recent surface water  
19 conditions (2006) with preindustrial conditions (i.e. preacidification 1860). The results  
20 showed a marked increase in the number of acid impacted lakes, characterized as a  
21 decrease in ANC levels, since the onset of anthropogenic N and S deposition (**REA**  
22 **appendix 4 section 5**)

23 **Observed recent trends over time**

24 In the REA, more recent trends in ANC, over the period from 1990 to 2006, were  
25 assessed using monitoring data collected at the two case study areas. In both case study  
26 areas, nitrate and sulfate deposition decreased over this time period. In the Adirondack  
27 Mountains, this corresponded to a decreased concentration of nitrate and sulfate in the  
28 surface waters and an increase in ANC (**REA 4.2.4.2**). In the Shenandoah Mountains,  
29 there was a slight decrease in nitrate and sulfate concentration in surface waters  
30 corresponding to modest increase in ANC from 50 ueq/L in 1990 to 67 ueq/L in 2006  
31 (**REA 4.2.4.3 and REA appendix 4 section 3.4**).

32

1 **Critical loads**

2 In the REA, the quantified relationship between deposition and ANC was  
3 investigated using ecosystem acidification models, also referred to as **acid balance**  
4 **models or critical loads models (REA Chapter 4 and REA Appendix 4)**. These  
5 models quantify the relationship between deposition of N and S and the ability of an  
6 ecosystem to counterbalance or buffer the deposition. The ecosystem acidification  
7 models simulate a variety of water and soil acidification responses at the laboratory, plot,  
8 hillslope, and catchment scales. For example, the level of deposition that causes a  
9 specified level of an ecosystem endpoint could be calculated (e.g. a critical load for  
10  $ANC=50\mu eq/L$ ). A summary of acidification models is given in ISA appendix A and  
11 further discussed in section 5.3.2.3. In the REA analysis, critical loads and their  
12 exceedances were calculated for four values of ANC (i.e., ANC of 0, 20, 50, and 100  
13  $\mu eq/L$ ) for 169 lakes in the Adirondack Mountains and 60 streams in the Shenandoah  
14 Mountains.

15 In summary, EPA staff concludes from the REA analysis, in combination with  
16 information presented in the ISA, that a quantitative relationship exists between the level  
17 of surface water ANC and an amount of nitrogen and sulfur deposition. These  
18 relationships are shown by long-term trends going back to preindustrial conditions in the  
19 1860s, recent trends since the 1990s and critical loads modeling.

20  
21 **5.3.2.3 Relating the ecological indicator to deposition: *How do steady state models***  
22 ***compare to dynamic models?***  
23

24 Models are important tools to evaluate how multiple environmental factors alter  
25 the relationship between ANC atmospheric deposition. There are two general types of  
26 acidification models: steady-state and dynamic. These models make different  
27 assumptions, indicate different time horizons for their critical loads and they have  
28 different data requirements.

29  
30 ***Basic approach of steady-state vs. dynamic acidification models***

31 The basic principle of the steady-state approach of aquatic acidification models is  
32 to determine the maximum acid input that will result in adequate biogeochemical

1 conditions to sustain ecosystem health. Adequate biogeochemical conditions is a subjective  
2 term that relates to a particular benchmark (e.g. ANC = 20, 50, 100), representing  
3 different degrees of protection of aquatic ecosystems against acidic deposition. The steady-  
4 state models relate an aquatic ecosystem's critical load to the weathering rate of its  
5 drainage basin expressed in terms of the base cation flux. The weathering of soil minerals  
6 is often a major source of base cation supply to an ecosystem. It is considered one of the  
7 governing factors of ecosystem critical loads.

8 A dynamic model includes mathematical descriptions of processes that are  
9 important in controlling the chemical response of a catchment. One of the most well-  
10 known dynamic models of aquatic acidification is MAGIC (Cosby et al., 1985a; 1985b;  
11 1985c). It is a lumped-parameter model of soil and surface water acidification in  
12 response to atmospheric deposition based on process-level information about  
13 acidification. "Lumped-parameter" refers to the extent that spatially distributed physical  
14 and chemical processes in the catchment are averaged or lumped together without  
15 affecting the model's reproduction of catchment response. Process-level information  
16 refers to how the model characterizes acidification into (1) a section in which the  
17 concentrations of major ions are assumed to be governed by simultaneous reactions  
18 involving  $\text{SO}_4^{2-}$  adsorption, cation exchange, dissolution-precipitation- speciation of  
19 aluminum, and dissolution-speciation of inorganic carbon; and (2) a mass balance section  
20 in which the flux of major ions to and from the soil is assumed to be controlled by  
21 atmospheric inputs, chemical weathering, net uptake and loss in biomass and losses to  
22 runoff. At the heart of MAGIC is the size of the pool of exchangeable base cations in the  
23 soil. As the fluxes to and from this pool change over time owing to changes in  
24 atmospheric deposition, the chemical equilibria between soil and soil solution shift to  
25 give changes in surface water chemistry. The degree and rate of change of surface water  
26 acidity thus depend both on flux factors and the inherent characteristics of the affected  
27 soils.

28  
29 *Trajectory of recovery for ecosystems from CL calculated by steady-state vs. dynamic*  
30 *acidification models*

1           Steady-state models assume that the ecosystem is in equilibrium with the critical  
2 load of deposition; therefore the long-term sustainable deposition is indicated. This is the  
3 relevant information needed to provide protection from deposition in perpetuity as the  
4 system comes into equilibrium with the pollutant critical load (**ISA Appendix D**). In the  
5 U.S., few (if any) ecosystems qualify as steady-state systems. Therefore the assumption  
6 of equilibrium in the steady-state model is often false. This has implications for the  
7 temporal aspects of ecosystem recovery. The steady-state models give no information  
8 concerning the time to achieve the equilibrium or what may happen to the receptor along  
9 the path to equilibrium. The recovery of an ecosystem based on a critical load from a  
10 steady state model may take several hundred years. In other words the assumption that  
11 attainment of a deposition values below the steady-state critical load will result in  
12 biological recovery within a specified time period may not be valid.

13           Dynamic models calculate time-dependent critical loads and therefore do not  
14 assume an ecosystem is in equilibrium. This is the relevant information needed to provide  
15 protection from damage by the pollutant within a specified time frame. As a general rule,  
16 the shorter the time frame selected, the lower the critical load.

17           The most comprehensive study done in the United States is Holdren et al. 1992  
18 that compared critical loads calculated by the dynamic MAGIC model versus SSWC  
19 steady-state approach. A 50-yr simulation critical load was obtained from the MAGIC  
20 model. Holdren et al. 1992 found that both models yielded the same general trends. The  
21 critical load estimates projected using the dynamic versus steady-state models are  
22 consistently higher. Both model produced critical load values approximately equal for  
23 systems with critical loads of about zero. However, at higher critical load values the two  
24 model outputs diverge rapidly, implying that watersheds with larger inherent buffering  
25 capacities respond more slowly to a given level of acidic deposition. The apparent  
26 reason for this is that the watersheds represented by the dynamic model retain a larger  
27 fraction of their buffering capacity in the base cation exchange pool for the 50-yr time  
28 scale of the simulation. In the Steady-state models, the cation exchange pool is assumed  
29 to be in equilibrium and does not provide additional buffering be on what results during  
30 equilibrium conditions.

31

1 ***Data Requirements of steady-state vs. dynamic acidification models***

2 There are various factors that modify the ANC to deposition relationship, which  
3 are described by models that parameterize ecosystems to simulate the process of  
4 acidification. The steady-state models used for critical loads analysis in the REA  
5 required input data for between 17 and 20 variables, including water chemistry data from  
6 the TIME and LTM programs, which are part of the Environmental Monitoring and  
7 Assessment Program (EMAP). A summary of the variables for steady state models (and  
8 data sources for the calculations made in the REA) is given in Appendix A.

9 The data requirements required to run dynamic models, such as MAGIC, are  
10 greater. The equations that characterize the chemical composition of soil water in  
11 MAGIC contain 33 variables and 21 parameters (Cosby et al. 1985). Data required to  
12 conduct dynamic modeling are not available for as many places as the data required to  
13 conduct steady-state modeling.

14  
15 ***Comparison of two steady-state models: FAB and SSWC***

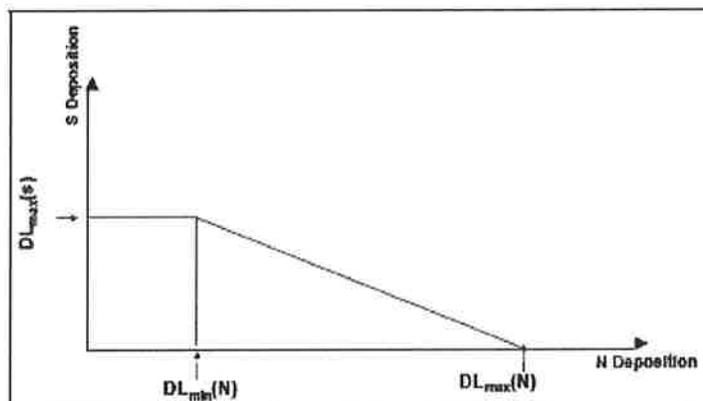
16 The steady state models used in the REA were the Steady State Water Chemistry  
17 model (SSWC), and the First-order Acid Balance model (FAB). The SSWC and FAB  
18 models were used to calculate critical loads for specified ANC levels in the case study  
19 areas.

20 The SSWC and FAB make different assumptions of ecosystem function. Most  
21 notably, biogeochemical pathways of N deposition are considered differently in the two  
22 models. In the SSWC model, sulfate is assumed to be a mobile anion (i.e. S leaching = S  
23 deposition), while nitrogen is retained in the catchment by various processes. The  
24 assumption that all N is retained by the ecosystem and does not contribute to acidification  
25 is incorrect because in many ecosystems nitrate leaching is observed. If nitrate is leaching  
26 out of an ecosystem, it cannot also be true that it has all been retained. Nitrate leaching is  
27 determined from the sum of the measured concentrations of nitrate in the runoff. The  
28 critical load for sulfur that is calculated by SSWC can be corrected for the amount of  
29 nitrogen that contributes to acidification. When an exceedence value for the critical load  
30 is calculated, the critical load is subtracted from S deposition plus the amount of nitrate

1 leaching, as it represents the difference between N deposition and N retention by the  
2 ecosystem. N leaching data used in this calculation are considered robust.

3 In contrast to the SSWC approach, the FAB model includes more explicit  
4 modeling of N processes including soil immobilization, denitrification, and wood  
5 removal, in-lake retention of N and S, as well as lake size. Although N cycling is more  
6 detailed in the FAB model, there is greater uncertainty in the input data needed to  
7 characterize the components of the N cycle. The FAB model yields a deposition load  
8 function for a specified level of an endpoint. This function is characterized by three  
9 nodes that are illustrated on Fig 5.5: 1. the maximum of amount of N deposition  
10 when S deposition equals zero ( $DL_{max}(N)$ ); 2. the amount of N deposition that  
11 will be captured by the ecosystem before it leaches ( $DL_{min}(N)$ ); and 3. the  
12 maximum amount of S deposition considering the N captured by the ecosystem  
13 ( $DL_{max}(S)$ ). The function represents many unique pairs of N and S deposition that  
14 will equal the critical load for acidifying deposition. The slope portion of the  
15 function will vary according to attributes of the water body that is modeled,  
16 including lake size and in-lake retention.

17



18

19 **Figure 5-5** Illustration of a generalized N + S deposition tradeoff curve that is  
20 calculated by using the FAB approach  
21

22 The EPA staff concludes that the available information supports using the steady-  
23 state acidification models to characterize the relationship between the ANC ecological  
24 indicator and total nitrogen and sulfur deposition. The steady state models take a simple

1 mass balance approach to characterizing ecosystem acidification and data is available for  
2 over 8,000 locations to conduct steady-state modeling.

3  
4 **5.3.2.4 Relating the ecological indicator to deposition:** *What is the appropriate*  
5 *ecosystem acidification model to represent the relationship between nitrogen and sulfur*  
6 *deposition, and the ecological indicator?*  
7

8 A combination of the SSWC and FAB models is proposed by EPA staff for  
9 catchment scale modeling for use in developing critical loads to specify the form of the  
10 standards. A modified SSWC model is used because there is high confidence in the  
11 availability and quality of the input data that is required by this model. The SSWC model  
12 for aquatic acidification is expressed as equation 1.

13  
14 
$$CL_{ANC\lim}(N + S) = ([BC]_0^* - [ANC_{\lim}])Q \quad (1)$$

15  
16 Where,

17  $CL_{ANC\lim}(N+S)$  = depositional load of S and N that does not cause the ecosystems to  
18 exceed a given  $ANC_{\lim}$  (meq/m<sup>2</sup>/yr)

19  $[BC]_0^*$  = the preindustrial concentration of base cations (μeq/L)

20  $ANC_{\lim}$  = a ANC limit (μeq/L)

21  $Q$  = surface water runoff (m/yr) (this is typically equal to precipitation –  
22 evapotranspiration)

23  
24 This model is further constrained by a quantity of N which would be taken up,  
25 immobilized or denitrified by ecosystems and used to adjust the quantity of deposition  
26 required to meet a specified critical load. This term is represented as  $CL_{min}(N)$  in the  
27 FAB model and illustrated in Fig. 5.5. For application in the form of the standard and in  
28 the following discussion, the parameter is designated with the abbreviation  $N_{eco}$ . The  
29 acid-base model constrained by  $N_{eco}$  is expressed by equation 2.

30  
31 
$$CL_{ANC\lim}(N + S) = ([BC]_0^* - [ANC_{\lim}])Q + N_{eco} \quad (2)$$

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Where,

$N_{eco}$  = nitrogen retention and denitrification by terrestrial catchment and nitrogen retention in the lake

Two options to derive the term  $N_{eco}$  were considered, each yielding different results. The first approach is to calculate the long-term amount of N an ecosystem can immobilize and denitrify before leaching (i.e. N saturation) that is derived from the FAB model [denoted as DLmin(N) in the FAB model]. This approach requires the input of multiple ecosystem parameters and is expressed by eq 3.

$$N_{eco} = fN_{upt} + N_{ret} + (1 - r)(N_{imm} + N_{den}) \quad (3)$$

Where,

$N_{upt}$  = nitrogen uptake by the catchment (meq/m<sup>2</sup>/yr)

$N_{imm}$  = nitrogen immobilization by the catchment (meq/m<sup>2</sup>/yr)

$N_{den}$  = denitrification of nitrogen in the catchment (meq/m<sup>2</sup>/yr)

$N_{ret}$  = in-lake retention of nitrogen (meq/m<sup>2</sup>/yr)

f = forest cover in the catchment (dimensionless parameter)

r = fraction lake/catchment ratio (dimensionless parameter)

The second approach for estimating  $N_{eco}$  is to take the difference between N deposition and measured N leaching in a catchment as expressed by eq 4.

$$N_{eco} = Dep_N^{Total} - N_{leach} \quad (4)$$

CASAC has advised EPA to use the second approach to derive the  $N_{eco}$  term:

“In principal, Equation 3, captures many of the major landscape and ecological factors that influence the processing of atmospherically deposited N within an ecosystem, including biological and abiotic retention of N (immobilization, uptake and sedimentation), and gaseous loss (denitrification) after N has been deposited and/or transported. This approach opens the “black box” and attempts to estimate some of the component parts of N processing and loss. It is best modeled as a dynamic process because such factors as age

1 and stage of vegetation, soil moisture, moisture regime, and nutrient demand  
2 will affect various components of N cycling. For many ecosystems, complete  
3 supporting data may not be available.

4 Equation 4 is basically a mass balance approach that keeps track of  
5 inputs and outputs. It does not include the detailed biogeochemical processes  
6 identified in Equation 3. Data for this latter approach may be much more  
7 readily available than the approach using Equation 3. Watershed N retention  
8 can be estimated using this approach (e.g., Lovett et al. 2000, and many  
9 others), but the processes involved in the retention are not detailed. This  
10 approach is most effectively used when hydrologic boundaries (e.g.,  
11 watershed) are defined. Hence, the Panel recommends that the mass-balanced  
12 (i.e., Equation 4) approach should be used.” (CASAC, April 29, 2010)  
13

14 (A comparison of calculating Neco using both methods is provided in Appendix A)  
15

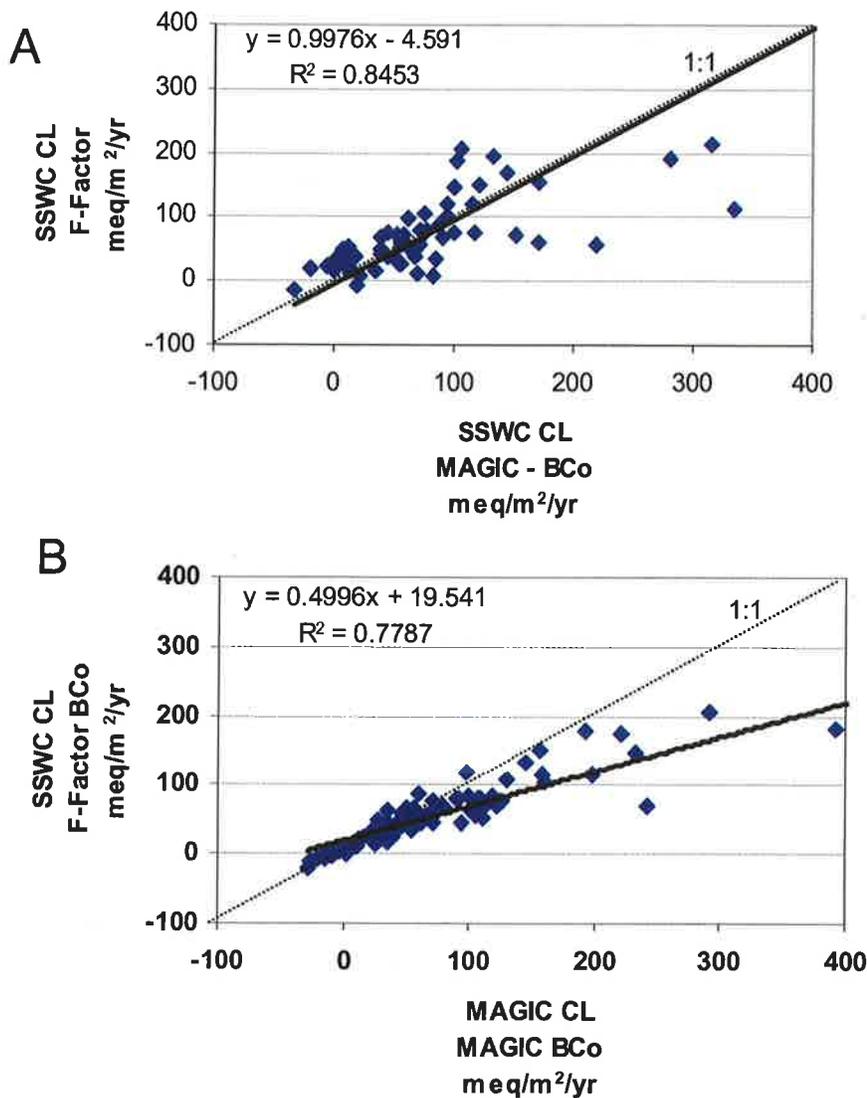
### 16 ***Two methods to calculate pre-industrial base cation weathering: F factor and MAGIC***

17 The preindustrial concentration of base cations ( $[BC]_0^*$ ) is calculated to represent  
18 conditions prior to industrialization (~about 1860). It incorporates the main source of  
19 base cations to an ecosystem including preindustrial weathering from soil and pre-  
20 industrial base cation deposition. It is therefore considered one of the governing factors  
21 of critical loads.  $[BC]_0^*$  is commonly calculated using one of two approaches: dynamic  
22 modeling (i.e. MAGIC) and calculation by the F-factor approach (Henriksen and Posch  
23 2001). Staff propose that the base cation weathering rates may be calculated by either  
24 method.

25 In this section, critical load estimates obtained from two steady-state approaches  
26 were compared. The exercise is not intended to provide an assessment of the accuracy of  
27 the two models, but rather to provide a means for evaluating the relative performance of  
28 the two different models. The EPA conducted analysis compared steady-state CL values  
29 based on Henriksen and Posch (2001) F-factor approach and output from the MAGIC  
30 model. The primary purpose of the F-factor is to obtain estimates of preindustrial surface  
31 water base cation concentrations ( $[BC]_0^*$ ) for equation 1. The MAGIC (Cosby et al.,  
32 1985) model can also be used to derive preindustrial surface water base cation  
33 concentrations. MAGIC is a process-based model designed to mimic the geochemical  
34 effects of mineral weathering, soil cation exchange, and other watershed processes. Once  
35 the model has been calibrated for a watershed, it can be run to simulate how surface

1 water chemistry changes with time and to predict preindustrial cation concentrations  
2 ( $[BC]_0^*$ ) to be used in the steady-state SSWC CL model in equation 1.

3 The comparison of CLs between the F-factor and MAGIC approached was done  
4 for two regions, streams in Southern Appalachia and lakes in the Adirondack Mountains.  
5 For 67 streams and 99 lakes,  $[BC]_0^*$  were determined for both approaches and CLs were  
6 calculated using the same value of Q. The results are show in Fig 5.6. For this analysis,  
7 the steady-state MAGIC model yielded critical load values that show the same trend for  
8 both regions, but were on average 16 meq S /m<sup>2</sup>-yr for the Adirondack Lakes and 5 meq  
9 S/m<sup>2</sup>-yr for Southern Appalachia streams higher than those from the SSWC F-factor  
10 approach. The two models converge at low critical, but diverge as the buffering potential  
11 for watersheds increase. This is particularly the case for CL values above 80 meq/m<sup>2</sup>-  
12 yr for lakes in the Adirondack Mountains. These results are consistent with similar  
13 comparison of critical load done by Holdren et al. 1992. Holdren et al 1992  
14 found on average that the MAGIC model yielded CL values that were on average  
15 29 meq S /m<sup>2</sup>-yr higher than those from the SSWC model. Holdren et al 1992 also  
16 found that as the buffering potential for watersheds increased, as indicated by increasing  
17 CLs, the results from the two models gradually diverge.



1  
2  
3 **Figure 5-6 Relationship between CLs using pre-industrial base cation**  
4 **weathering ( $[BC]_0^* = BC_0$ ) calculated by MAGIC versus the F-factor**  
5 **methods: A) 67 streams in the Southern Appalachian and B) 99 lakes in**  
6 **the Adirondacks Mountains.**

7  
8  
9 **5.3.2.5 Relating the ecological indicator to deposition:** *How can we aggregate critical*  
10 *loads, derived from catchment-scale acidification models for a target ANC, across*  
11 *broader areas to develop a deposition metric that adequately for a nationally*  
12 *representative acid-sensitivity category?*

13  
14 Acidification models are considered the best way to describe the relationship between  
15 ANC and deposition. It is important to emphasize that the acidification models are only

1 applied at the spatial scale of the catchment. Response to N and S deposition will vary  
2 among catchments. However, modeling every catchment in a region (i.e. a spatial area  
3 that includes a large population of individual catchments) is implausible at this time due  
4 to the data limitations. A method to extrapolate watershed-scale analysis to a region was  
5 developed in the REA. In that method, the critical loads (combined N+S load for a  
6 selected ANC level) developed for the case study sites were applied over a region using  
7 water chemistry data. Critical load exceedance (i.e., the amount of actual deposition  
8 above the critical load, if any) was calculated for each waterbody in the region to quantify  
9 the number of lakes or streams that receive levels of deposition exceeding the CL. Lakes  
10 and streams with positive exceedance values, where actual deposition was above its  
11 critical load, were not protected at that critical limit (e.g. ANC= 20, 50, 100  $\mu\text{eq/L}$ ; **REA**  
12 **Appendix 4**).

13 The relationship between ANC and N and S deposition is also applicable  
14 nationally. Areas that have similar geologic underpinnings, mineral weathering rates, and  
15 hydrology should show similar sensitivity to  $\text{NO}_y$  and  $\text{SO}_x$  deposition. Weathering rate  
16 of geologic parent material is the main source of base cations to an ecosystem, and it is  
17 therefore considered one of the governing factors of ecosystem critical loads. Landscape  
18 features that are linked to ecosystem acid-sensitivity include lithology, elevation, percent  
19 forested watershed, and watershed area (Sullivan et al. 2007).

20 How do we use catchment scale acidification modeling to inform the NAAQS? The  
21 goal is to have a national applicable standard, but still take into account catchment base  
22 processes. As previously noted, acidification models are parameterized for catchments  
23 and their critical loads vary at the spatial scale of the catchment. However, aggregating  
24 critical loads from multiple catchments will allow for an appropriately representative  
25 deposition value, which provides adequate protection of ecosystems and can be applied  
26 over larger spatial areas. For this reason, EPA Staff proposes evaluating a population of  
27 waterbodies that are sensitive to acidification to calculate a benchmark depositional load  
28 (called the deposition metric) in which a specified percentage of the population does not  
29 exceed their critical load for a selected ANC limit. This approach uses the distribution of  
30 critical loads from a population to derive a value that is intended to provide protection

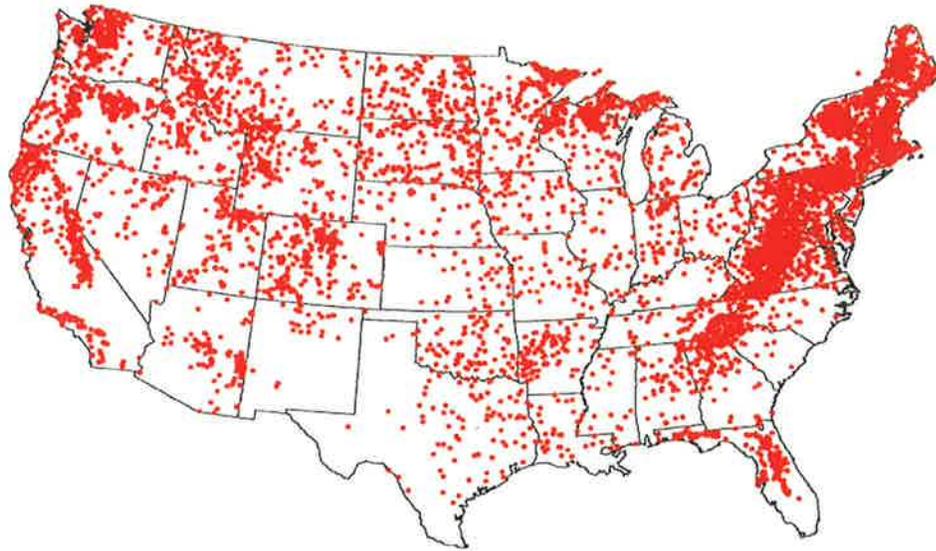
1 over a spatial area that is larger than the individual catchment for which a single critical  
2 load may be calculated. The deposition metric is expressed by the following equation:

$$DL_{\%ECO}(N + S) = \left\{ \left( [BC]_O^* - [ANC_{lim}] \right) Q \right\}_{\%ECO} \quad (5)$$

3  
4  
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6  
7 The change from the CL term to the DL term reflects the change from a critical load  
8 for deposition to a specific catchment to a deposition load that applies across a population  
9 of catchments in a broader area.  $N_{ECO}$  would then be added to  $DL_{\%ECO}$  to calculate the N  
10 and  $SO_2 + SO_4^{-2}$  tradeoff curves (section 5.3.2.8)

11  
12 **5.3.2.6 Deposition metric:** *What critical load data is available and what populations of*  
13 *catchments are excluded? For any acid sensitive category, how is the distribution*  
14 *of critical loads evaluated in developing the deposition metric?*

15  
16 Steady state critical loads calculated for ANC that were used in this analysis were  
17 either previously published in the literature or calculated using the SSWC method  
18 from water quality data collected by national monitoring programs. A map of critical  
19 loads is presented in Fig. 5.7.



1  
2 **Figure 5-7 Sites of CLs calculated by SSWC that are used in the Policy**  
3 **Assessment analysis**  
4

5 To ensure the population of water bodies included in the analysis were those  
6 sensitive to acidity caused by atmospheric deposition, several criteria were applied to the  
7 CL dataset to remove watersheds in which organic acids, acid mine drainage or naturally  
8 low base cation weathering caused acidification.

- 9
- 10 ■ The pre-industrial ANC was calculated for each site in which there was  
11 sufficient data. Certain pre-industrial ANC values, defined as  $ANC < \text{the target ANC}$  ( e.g.  $50 \mu\text{eq/L}$ ), indicate natural acidity due to naturally low BC  
12 weathering and were removed from the data set.
  - 13 ■ All  $CL \leq 10 \text{ meq/m}^2/\text{yr}$  were removed. This is a second screen to remove  
14 catchments that may be naturally acidic for which pre-industrial ANC values  
15 could not be calculated.
  - 16 ■ To remove catchments that are dominated by acidity caused by acid mine  
17 drainage, if the concentration of  $\text{SO}_4^{2-}$  of the water was  $>400 \text{ ueq/L}$  twice or

1 more than expected by S deposition the catchment was considered to be  
2 dominated by acid-mine drainage and removed from the dataset.

- 3 ■ The cause of aquatic acidity was considered to be dominated by organic acids  
4 if the concentration of DOC in the water was > 10mg/L, sites exceeding 10  
5 mg/L were excluded from the dataset.

6  
7 The latitude and longitude of each site was used to determine its membership to  
8 an acid-sensitive category. Multiple methods for defining an acid sensitive category are  
9 presented in section 5.3.2.7. The distribution of critical loads from a population that  
10 represents an acid-sensitivity category was then evaluated. A deposition value that  
11 protects a specified percentage of the ecosystems (called the deposition metric) would be  
12 selected. Each acid-sensitivity category would have a deposition metric that protects the  
13 same percentage of ecosystems (see Section 5.4-5.6 for a comprehensive discussion of  
14 the levels). The deposition metric would be a value representing a specified percentile of  
15 the distribution, such as the 95<sup>th</sup> percentile. The distributions of critical loads for many  
16 definitions of acid sensitive areas are skewed to the right, which implies that a central  
17 estimate using the mean, would not be projected to achieve the target ANC of the  
18 majority of acid-sensitive ecosystems; therefore it may be preferable to calculate the  
19 deposition metric to target protecting a higher percentage of catchments. For example, if  
20 the 75% or 90% of the aquatic ecosystems are targeted for protection, then the deposition  
21 metric that represents the critical load for the 75<sup>h</sup> or 90<sup>th</sup> percentile of the population  
22 would be selected.

23  
24 **5.3.2.7 Landscape categorization:** *How is the U.S. landscape categorized based on acid-*  
25 *sensitivity?*

26 Two general approaches are suggested. The first approach is to have one  
27 population of water bodies represent the entire nation and select a deposition metric from  
28 this one large population. In this approach the U.S is not subdivided into acid-sensitivity  
29 categories. The second approach is to subdivide the U.S. into acid-sensitivity categories  
30 based on criteria that are known to be associated with acid-sensitivity and for which  
31 national-scale data sets are available. A population of catchments could then be

1 identified to represent each acid-sensitive category. Several categorization possibilities  
 2 are discussed under the second approach.

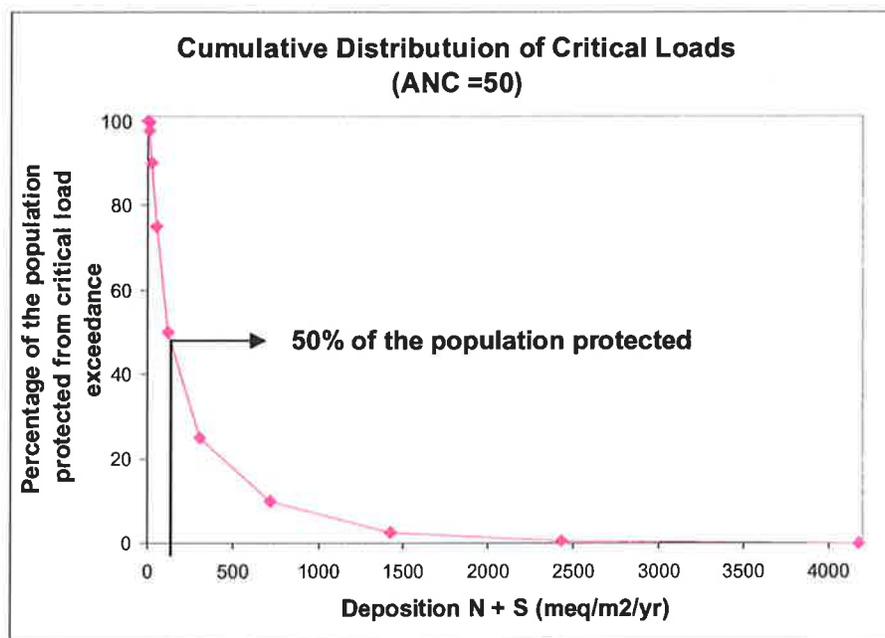
3  
 4 **Option 1: No subdivision of the U.S.**

5 Option 1 is a simple approach in which the U.S. is not subdivided into acid-  
 6 sensitivity categories and the total population of critical loads represents the entirety of  
 7 the U.S. The deposition metric would be selected by evaluating the distribution of CL  
 8 from the entire population after screening out preindustrial ANC < 50  $\mu\text{eq/L}$  and CL < 10  
 9  $\text{meq/m}^2/\text{yr}$ , as described in section 5.3.2.6. Deposition metrics that protect 90%, 75% and  
 10 50% of the population are given as examples in Table 5.1. Figure 5.8 shows the  
 11 cumulative distribution of critical loads of waterbodies at ANC=50  $\mu\text{eq/L}$ . Note that for  
 12 an ANC=50  $\mu\text{eq/L}$  the greatest CL is 4179  $\text{meq/m}^2/\text{yr}$ , while 50% of the waterbodies have  
 13 CLs less than 118  $\text{meq/m}^2/\text{yr}$ , thus indicating the skewed distribution of the data set.

14  
**Table 5.1. Descriptive statistics of the national CL data set.**

AN C $\mu\text{eq/L}$	# CL	Mean Nec o	After excluding sites for sulfate > 40 0, DOC > 10, pre- industrial ANC < target ANC, then excluding sites with CL < 10, the resulting CL in analysis n=	CL mean ( $\text{meq/m}^2/\text{yr}$ )	C L St er	Example deposition metrics		
						DL %90 ( $\text{meq/m}^2/\text{yr}$ )	DL %75 ( $\text{meq/m}^2/\text{yr}$ )	DL %50 ( $\text{meq/m}^2/\text{yr}$ )
50	749 7	52	5280	257	5	27	55	118
20	749 7	52	5613	259	4	33	62	124

15  
 16



**Figure 5-8 Cumulative distribution of the critical loads for ANC =50  $\mu\text{eq/L}$  considered in the analysis (n =5281)**

The benefit to this approach is that it provides one deposition metric for the entire country, making it simple and easy to calculate and convey. The main weakness of this approach is the degree of uniformity in protection across the nation, as that it may over-protect areas of the nation that are least sensitive to acidification and under protect in areas that are most sensitive. The extent of possible over or under protected is discussed at the end of this section.

***Option 2: Acid-sensitivity categorization of the landscape***

In Option 2, four approaches to categorize the U.S based on criteria that influence acid-sensitivity are presented: 1) binary categorization (sensitive vs. less-sensitive), 2) 5 categories of sensitivity (based on cluster analysis), 3) one less-sensitive category and sensitive ecoregions considered individually and 4) all ecoregions considered individually. The extent to which current critical loads represent the categories as defined by each approach is discussed. Each approach presented uses the concept of ecoregions to define spatial boundaries of sensitive ecosystems and ANC to determine acid-

1 sensitivity. Numerous options were explored, but we have chosen to focus this  
2 discussion on those options recommended by the staff as most likely to be useful in  
3 setting the standards. Other parameters for which to base acid-sensitivity categorization  
4 that were explored include bedrock geology, soil base cation weathering, dissolved  
5 organic carbon and alkalinity. Staff suggests moving forward with ANC because it is the  
6 best indicator of acid-sensitivity integrating natural buffering capacity, historic load and  
7 current load, and there is good national coverage of ANC measurement from almost  
8 every ecoregions (level 3). A brief summary of ecoregions and ANC data that were used  
9 in the analysis is presented here, prior to the presentation of the options.

#### 10 Ecoregions

11 Ecoregions are “areas of similarity regarding patterns in the mosaic of biotic,  
12 abiotic, aquatic, and terrestrial ecosystem components, with humans being considered  
13 part of the biota” (McMahon and others 2001). EPA staff proposed considering  
14 Omernik’s ecoregions in this analysis of acid-sensitivity (maps may be downloaded at  
15 [www.epa.gov/wed/pages/ecoregions.htm](http://www.epa.gov/wed/pages/ecoregions.htm)). These ecoregions are categorized using a  
16 holistic; “weight-of-evidence” approach where the importance of factors may vary. The  
17 method used to map ecoregions is,

18 “based on the premise that ecological regions can be identified through  
19 the analysis of the patterns and the composition of biotic and abiotic phenomena  
20 that affect or reflect differences in ecosystem quality and integrity (Wiken 1986;  
21 Omernik 1987, 1995). These phenomena include geology, physiography,  
22 vegetation, climate, soils, land use, wildlife, and hydrology. The relative  
23 importance of each characteristic varies from one ecological region to another  
24 regardless of the hierarchical level”.

25 To evaluate the effectiveness of ecoregions, they must be tested against measures that  
26 relate to their purpose (Omernik 2004). The first publication of the ecoregions based on  
27 Omernik’s weight-of-evidence approach was published in 1987 (Omernik 1987). Current  
28 maps are refinements and revisions of the 1987 publication. Hierarchical levels were  
29 developed and a Roman numeral classification scheme was adopted to distinguish coarser  
30 (more general) and finer (more detailed) categorization. Level I is the coarsest level,  
31 dividing North America into 15 ecoregions (CEC 1997). At Level II, the continent is  
32 subdivided into 52 ecoregions (CEC 1997). Level III, is a further subdivision of Level II,

- 1 and divides N. America into 120+ categories. Level IV is a subdivision of Level III, it is
- 2 the finest scale of Omernik's ecoregions mapping and development of these maps is
- 3 currently in progress. Ecoregion level 3 are used here to categorize the landscape.

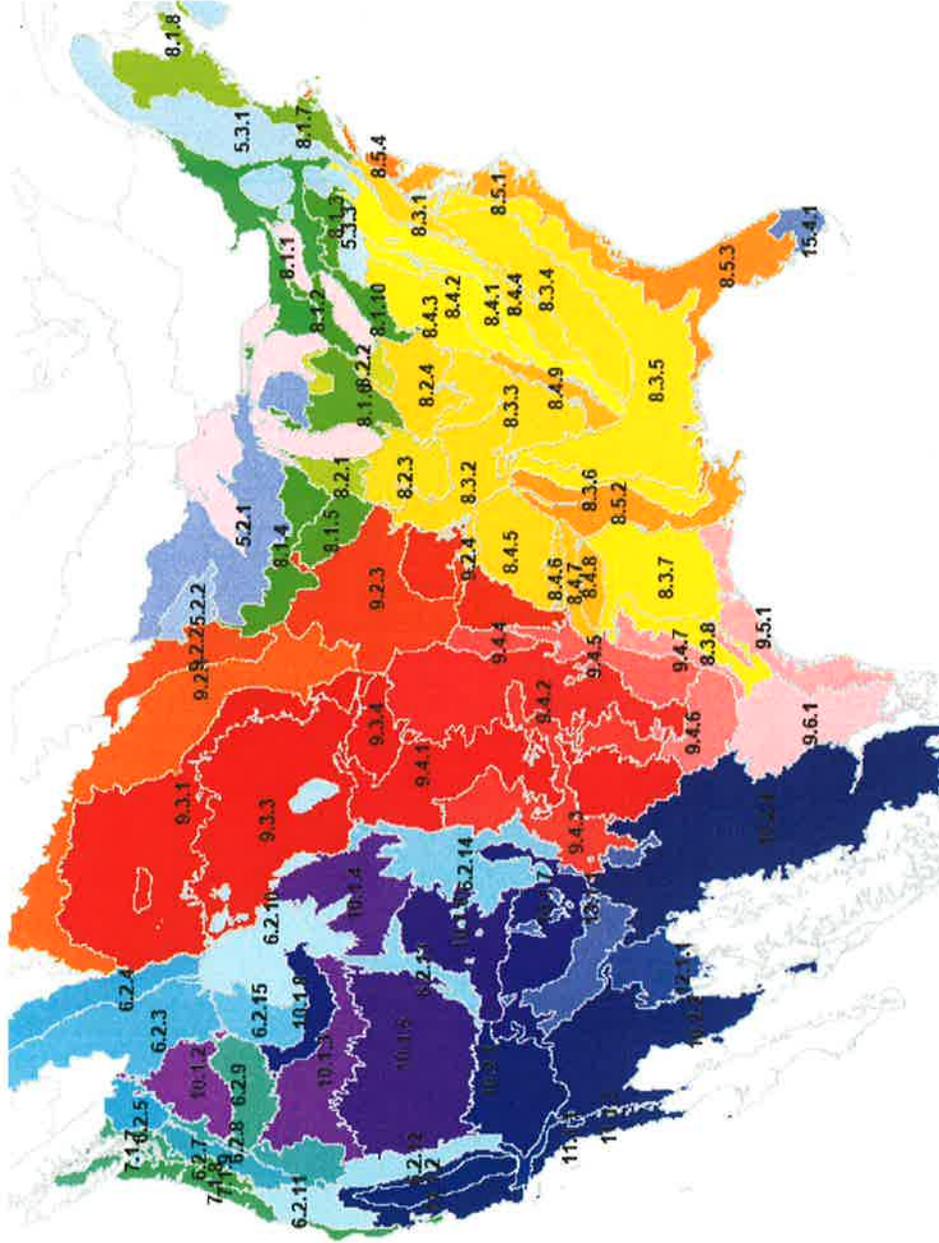


Figure 5-9 Map that illustrates Omernick's ecoregions (level 3).

1 ANC data

2 Water chemistry data on ANC was collected from several national monitoring networks. The  
 3 data sources are summarized on Table 5.2. These data sources included approximately 11,000  
 4 observations. For sites with more than one year of data, the measurements were averaged over  
 5 the past 5 years. For example, if data for 2008, 2007, 2006, 2005, 2004, and 2003 were  
 6 available, then the years from 2004-2008 were averaged. If it had data from 1990 and 2002 they  
 7 were not averaged because the 1990 point was beyond the cutoff date.

8

**Table 5-2 Summary of data sources considered for the evaluation of national ANC**

<b>Program</b>	<b>Dates of observations</b>	<b>Reference</b>
EPA Long Term Monitoring Vermont (LTM_VT)	1983-2007	EPA/903/R-00/015
EPA Eastern Lakes Survey (ELS)	1984	EPA/620/R-93/009
Adirondack Lake Survey (ALS)	1984-1987	Stoddard.et.al.WRR.1996
EPA Western Lake Survey (WLS)	1985	EPA 620-R-05-005
EPA National Stream Survey (NSS)	1986	Stoddard.et.al.WRR.1996
VT SSS	1987 & 2000	EPA 841-B-06-002
EPA Long Term Monitoring_Colorado sites (LTM_CO)	1990-1994	Stoddard.et.al.WRR.1996
EPA Long Term Monitoring_Midwest Sites (LTM_MW)	1990-2000	Stoddard.et.al.WRR.1996
VT SSS LTM	1990-2007	Stoddard.et.al.WRR.1996
EPA Long Term Monitoring_Pennsylvania sites(LTM_PA)	1990-2007	Stoddard.et.al.WRR.1996
EPA Long Term Monitoring_Catskill sites (LTM_CAT)	1990-2007	EPA 905-R-92-001
EPA Long Term Monitoring: Annual average from 1992-2007	1990-2007	EPA/600/4-88/032
EPA EMAP Northeast Lake Survey	1991-1994	Stoddard.et.al.WRR.1996
EPA Long Term Monitoring_Maine sites (LTM_ME)	1992-2007	reg1_qa.pdf
Regional Environmental Monitoring Program_Maine sites (REMAP_ME)	1993	Stoddard.et.al.WRR.1996
EPA EMAP_Mid Atlantic streams (EPA EMAP_MAIA)	1993-1996	EPA/R-06/XX
EPA EMAP_Mid Atlantic streams (EPA	1997-1998	EPA-600-388-021a

<b>Program</b>	<b>Dates of observations</b>	<b>Reference</b>
EMAP MAIA )		
EPA EMAP Western Stream and River Survey (EMAP WEST )	2000-2004	EPA/600/3-86/054b
EPA National Lakes Survey (NLS)	2010	EPA 841-F-09-007.
USGS NAWQA Program		<a href="http://water.usgs.gov/nawqa/">http://water.usgs.gov/nawqa/</a>
EPA Storet Program		<a href="http://www.epa.gov/storet/">http://www.epa.gov/storet/</a>

1

2 ***Option 2a: Categorize the U.S. into 2 categories (sensitive or less--sensitive) based on ANC***  
3 ***and ecoregions***

4 In this approach, ANC, considered the best indicator of acidification, is used to determine  
5 which ecoregions in the U.S. are sensitive to acidification. All ecoregions in the U.S. are then  
6 categorized as either sensitive or less-sensitive. A deposition metric is calculated for each  
7 category. The data sources used in this analysis are described first, followed by a more detailed  
8 description of the analysis.

9

10 *Defining acid-sensitivity of ecoregions based on ANC*

11 This analysis compared whether mean ANC differed between ecoregions when the entire  
12 dataset versus a subset of the dataset containing observations that ranged from -50 to 200  $\mu\text{eq/L}$   
13 was used in the analysis. The sample size and mean ANC value for each ecoregion is presented  
14 in table 5.3. In the data subset, the values ranging from -50 to 200  $\mu\text{eq/L}$  were selected to focus  
15 on acid-sensitive areas, all ANC observations that were greater than 200  $\mu\text{eq/L}$  were excluded  
16 from the dataset because ANC above this level indicates that an eco-system is not very sensitive  
17 to acidification. All observation less than -50  $\mu\text{eq/L}$  were also deleted from the dataset. This is  
18 because ANC values below -50  $\mu\text{eq/L}$  are typically not the result of acidification resulting from  
19 deposition (Kauffman et al. 1992). Both analyses indicate that ecoregions 5, 6, 7 and 8 have the  
20 among the lowest mean ANC values compared to the other five ecoregions. After observations  
21 outside of the -50 to 200  $\mu\text{eq/L}$  are removed, ecoregions 12, 13, are 15 are represented by five or  
22 less observations, indicating there are few sites measured with ANC values within the range of  
23 acid-sensitivity due to deposition. Therefore ecoregions 5 through 11 are considered sensitive  
24 and all other ecoregions are considered less sensitive.

1 Next only those level three subdivisions of the ecoregions with 10 or more observations  
 2 between -50 to 200  $\mu\text{eq/L}$  were considered sensitive. Fig 5.10 lists the level three ecoregions  
 3 which are indicated to be sensitive by ANC data.

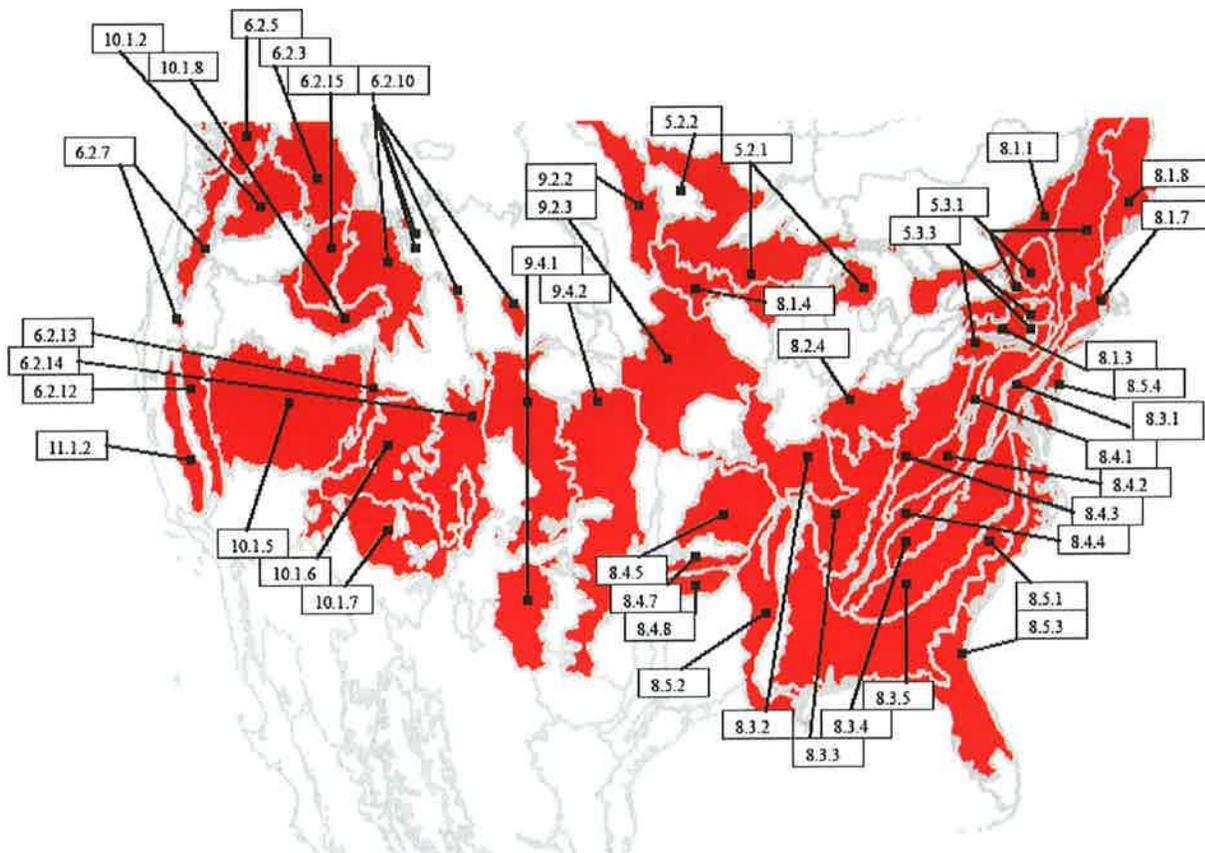
4

**Table 5-3 National ANC data. Observations from 6318 sites remained after filtering data to remove sites with an average value of ANC < -50 and > 200  $\mu\text{eq/L}$ . Color shaded boxes indicate ecoregions that are considered sensitive based on the ANC values. Kruskal-Wallis Test indicated ecoregions has a significant effect on ANC.**

<i>Ecoregion level 1</i>	<i>All ANC data</i>		<i>ANC data between -50 and 200 <math>\mu\text{eq/L}</math></i>	
	<i># sites</i>	<i>Mean ANC</i>	<i># sites</i>	<i>Mean ANC</i>
5	3519	212	2603	57
6	1952	892	754	90
7	177	775	30	68
8	5051	605	2680	77
9	1098	4694	94	149
10	592	2677	112	134
11	209	2663	35	93
12	19	2496	1	198
13	102	2869	3	146
15	12	406	5	164

5

6



1  
 2 **Figure 5-10 Map of sensitive ecoregions (red) using binary categorization approach**  
 3 **(sensitive vs. less-sensitive) with ANC observations between -50 and 200  $\mu\text{eq/L}$**   
 4  
 5

**Table 5-4. Descriptive statistics of the CL populations that result when the US is divided into two categories, sensitive and less-sensitive based on ANC data.**

ANC $\mu\text{eq/L}$	Sensitivity class	CL in class n=	Mean Neco	Neco St er	CL mean (meq/m <sup>2</sup> /yr)	CL St er	Example deposition metrics		
							DL %90 (meq/m <sup>2</sup> /yr)	DL %75 (meq/m <sup>2</sup> /yr)	DL %50 (meq/m <sup>2</sup> /yr)
50	sensitive	4553	55	0.4	219	4	26	51	106
50	less-sensitive	727	37	0.9	496	21	53	117	277
20	sensitive	4881	55	0.3	222	4	32	58	114
20	less-sensitive	732	37	0.9	508	21	63	122	291

1 Dividing the U.S. into two categories (sensitive and less-sensitive) provides a mechanism  
2 to reduce possible over-protection that may occur in less-sensitive regions via Option 1, which  
3 does not distinguish between areas that are sensitive and not-sensitive. In Option 2a, the  
4 sensitivity category is considered one area, in which one deposition metric could be calculated  
5 by evaluating all the CL data from the sensitive ecoregions. This would result in two deposition  
6 metrics for the U.S., one for sensitive areas and one for less-sensitive areas. The ecoregions in  
7 the sensitive category could be subdivided (See Option 2c).

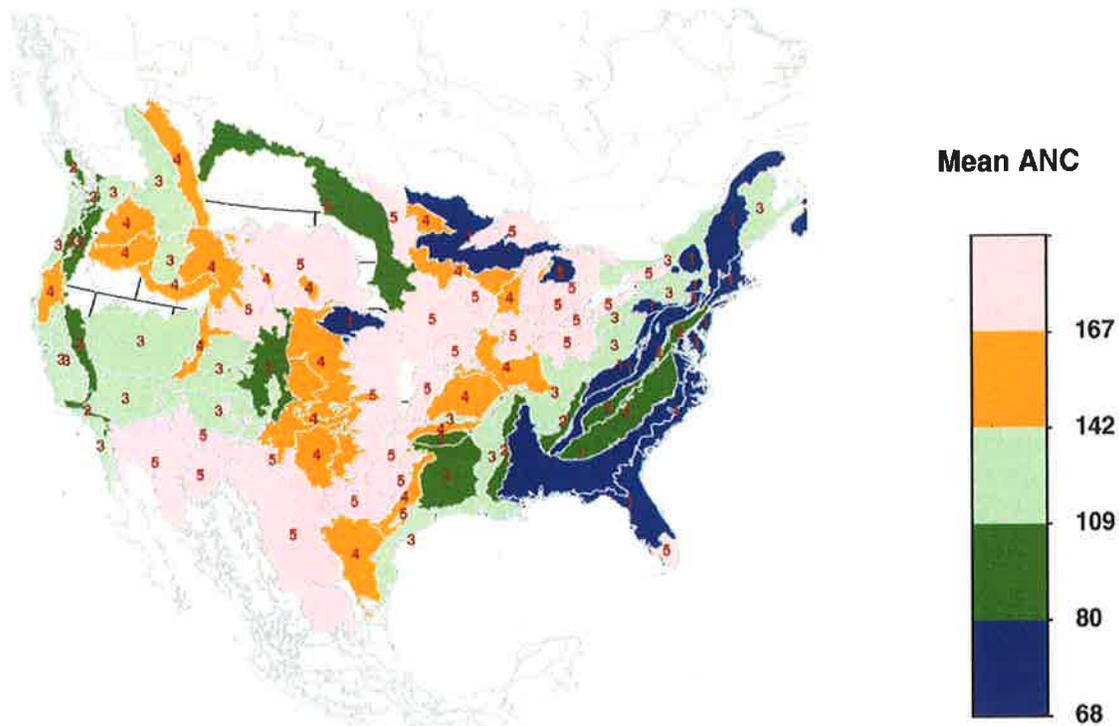
#### 8 9 **Option 2b: Cluster Analysis**

10 This approach uses ANC to categorize the acid-sensitivity of ecoregions based on a  
11 quantitative cluster analysis. Additional analysis simultaneously considering multiple criteria  
12 including ALK, DOC and soil BC weathering datasets were considered, however they are not  
13 suggested mainly due to limitation in the national coverage of these datasets, and in the case of  
14 soil BC weathering, uncertainty in the method of calculation.

15 Cluster analysis is a method of grouping objects together based on a chosen similarity  
16 metric that is determined from a set of variables of interest. In this analysis ecoregions are  
17 grouped based on the distributions of ANC from waterbodies. Numerous algorithms exist for  
18 performing cluster analysis. We employed the *k*-means method, which involves assigning a pre-  
19 determined number of clusters (*n*) to the dataset and minimizing the Euclidean distances between  
20 the cluster centers and the watersheds. Distance in this case refers to the water quality variables,  
21 not geographic distance. The process was repeated for a number of different *n* values (2 to 40),  
22 and a combination of fit statistics and graphical examination of the resulting clusters was used to  
23 settle on a final solution. The 5 cluster solution was chosen in this way, which has an  $R^2$  of 0.71  
24 and is shown in Fig 5.11. In selecting the number of clusters, we balanced goals of a  
25 parsimonious number of clusters with ability of the clusters to explain a large portion of the  
26 variance in ANC across ecoregions.

27 The deposition metrics based on the CL ANC=50  $\mu\text{eq/L}$  are presented for each cluster in  
28 Table 5.5. In general the sensitivity indicated by the mean ANC level of the clusters matches the  
29 deposition metrics, with the exception of Cluster 3 and 4 which have deposition metrics  
30 indicating they should switch places in regards to sensitivity indicated by the deposition metrics  
31 of the 90<sup>th</sup>, 75<sup>th</sup> and 50<sup>th</sup> percentile. This reflects the skewed nature of the distributions of ANC  
32 within ecoregions, where ecoregions may be matched more closely on alternative percentiles of

1 the distribution than others. There are 5 ecoregions which do not have cluster membership  
 2 (Table 5.6); this indicates there was insufficient ANC data in these ecoregions for classification.  
 3 There are 8 ecoregions that do not have CL data, but do have cluster membership (Table 5.6.).  
 4 These ecoregions would be represented by the deposition metric in their cluster.  
 5



6 **Fig 5.11. Map of sensitive ecoregions based on clusters of log ANC values**  
 7 **between -50 and 200  $\mu\text{eq/L}$**

**Table 5-5. Descriptive statistics of the CL populations that result when the US is divided into 5 clusters based on log ANC values between -50 and 200  $\mu\text{eq/L}$ .**

Target ANC =	Sensitivity class (mean ANC)	CL n=	Mean Neco (meq/m <sup>2</sup> /yr)	Neco St er	CL mean (meq/m <sup>2</sup> /yr)	CL St er	Example deposition metrics		
							DL %90 (meq/m <sup>2</sup> /yr)	DL %75 (meq/m <sup>2</sup> /yr)	DL %50 (meq/m <sup>2</sup> /yr)
50	5 (167)	216	51	2	354	20	72	114	269
50	4 (142)	655	39	0.7	393	17	30	64	193
50	3 (109)	784	41	0.8	416	18	48	99	232
50	2 (80)	1113	52	1.0	183	6	26	52	111
50	1 (68)	2432	61	0.5	197	6	24	46	90

**Option 2c: One less-sensitivity category and individual sensitive ecoregions**

This option builds on option 2a. Like option 2a, the CLs from the ecoregions (level 3) considered less-sensitive would be combined into one population to represent the less-sensitive category. The ecoregions (level 3) considered sensitive using the ANC approach described in option 2a would be represented by the CLs only from the same ecoregions. There are 43 ecoregions (level 3) considered sensitive using option 2a. The deposition metrics of the sensitive ecoregions are presented in Table 5.6.

**Table 5-6. Descriptive statistics of the populations of CL ANC=50 µeq/L values that result for each ecoregion**

Sensitivity class		Eco-region level 3	Neco mean	CL N=	CL mean (meq/m <sup>2</sup> /yr)	CL St er	Example deposition metrics		
Cluster 5	AN C						DL %90 (meq/m <sup>2</sup> /yr)	DL %75 (meq/m <sup>2</sup> /yr)	DL %50 (meq/m <sup>2</sup> /yr)
4	X	10.1.2	28	22	177	40	26	37	122
na		10.1.3	37	41	95	18	18	27	52
5		10.1.4	19	16	244	47	34	50	258
3	X	10.1.5	27	43	266	53	16	41	107
3	X	10.1.6	36	5	106	37	22	23	119
3	X	10.1.7	23	3	74	8	61	61	71
4	X	10.1.8	39	7	158	58	16	73	134
3		10.2.1							
5		10.2.2	19	2	75	1	73	73	
5		10.2.4							
3		11.1.1	20	16	705	211	62	127	293
3	X	11.1.2	22	5	279	97	58	88	262
3		11.1.3	14	19	367	69	41	72	337
5		12.1.1	32	3	88	41	45	45	48
5		13.1.1	38	52	189	21	36	76	120
5		15.4.1							
1	X	5.2.1	45	469	158	10	16	26	56
4	x	5.2.2							
1	X	5.3.1	55	735	177	9	36	58	101
1	X	5.3.3	79	159	113	9	28	52	90
4	X	6.2.10	35	202	230	20	26	50	112
4		6.2.11	20	86	956	92	138	360	744
2	X	6.2.12	17	105	172	24	17	28	68
4	X	6.2.13	37	78	245	45	21	33	59
2	X	6.2.14	22	186	145	12	20	43	91

**Table 5-6. Descriptive statistics of the populations of CL ANC=50  $\mu\text{eq/L}$  values that result for each ecoregion**

Sensitivity class		Eco-region level 3	Neco mean	CL N=	CL mean (meq/m <sup>2</sup> /yr)	CL St er	Example deposition metrics		
Cluster 5	AN C						DL %90 (meq/m <sup>2</sup> /yr)	DL %75 (meq/m <sup>2</sup> /yr)	DL %50 (meq/m <sup>2</sup> /yr)
3	X	6.2.15	38	86	155	19	16	35	76
3	X	6.2.3	47	73	353	35	46	119	228
4		6.2.4	61	28	956	92	102	653	990
3	X	6.2.5	21	133	425	33	90	163	308
2	X	6.2.7	21	88	376	29	60	116	284
na		6.2.8	25	30	323	44	73	127	260
4		6.2.9	32	108	244	17	65	117	197
2		7.1.7	16	29	423	64	21	185	325
3		7.1.8	12	68	1098	100	273	368	945
2		7.1.9	4	11	662	100	260	357	523
3	X	8.1.1	72	43	716	102	133	280	541
3	X	8.1.3	72	88	255	20	104	144	216
na		8.1.2							
4	X	8.1.4	65	45	328	41	26	46	333
5		8.1.5	24	9	961	125	158	794	946
5		8.1.6	90	23	731	49	381	617	703
1	X	8.1.7	72	169	223	18	49	86	148
3	X	8.1.8	35	143	177	16	51	70	108
3		8.1.10	81	9	935	152	351	542	1011
4		8.2.1	77	3	667	101	482	482	687
5		8.2.2							
5		8.2.3							
5	X	8.2.4	104	9	892	76	642	674	937
2	X	8.3.1	50	41	306	33	113	138	218
5	X	8.3.2	82	12	659	132	329	338	466
3	X	8.3.3	67	17	1022	147	429	558	719
2	X	8.3.4	75	168	174	7	74	100	155
1	X	8.3.5	63	75	182	244	15	34	76
2		8.3.6	78	6	399	242	52	98	166
2		8.3.7	65	31	211	35	60	104	160
4		8.3.8	69	4	261	149	32	55	156
1	X	8.4.1	68	510	321	22	31	53	98
1	X	8.4.2	82	200	160	16	24	36	82
3	X	8.4.3	85	19	346	59	119	168	316
2	X	8.4.4	78	379	98	5	21	38	68
4	X	8.4.5	70	24	938	96	243	642	907
3		8.4.6	73	14	102	28	25	42	67
4	X	8.4.7	67	23	129	22	33	64	94

**Table 5-6. Descriptive statistics of the populations of CL ANC=50 µeq/L values that result for each ecoregion**

Sensitivity class		Eco-region level 3	Neco mean	CL N=	CL mean (meq/m <sup>2</sup> /yr)	CL St er	Example deposition metrics		
Cluster 5	AN C						DL %90 (meq/m <sup>2</sup> /yr)	DL %75 (meq/m <sup>2</sup> /yr)	DL %50 (meq/m <sup>2</sup> /yr)
2	X	8.4.8	68	25	141	28	30	65	94
2		8.4.9	61	23	309	82	53	101	167
1	X	8.5.1	44	16	159	39	111	121	245
3		8.5.2	73	13	942	159	223	461	987
1	X	8.5.3	59	53	116	25	13	25	52
1	X	8.5.4	64	43	62	6	22	33	59
2		9.2.1	53	2	122	24	98	98	122
5	x	9.2.2							
5	X	9.2.3	21	13	425	40	249	337	392
5		9.2.4	72	16	365	26	190	309	361
na		9.3.1	55	2	211	101	110	110	211
5		9.3.3	32	21	278	73	78	106	139
1		9.3.4	38	3	79	15	49	49	87
4	X	9.4.1	38	11	30	3	17	20	27
5	X	9.4.2	59	15	148	18	58	102	138
4		9.4.3	30	2	36	6	30	30	36
?		9.4.4	66	6	419	55	268	315	398
5		9.4.5	69	28	257	21	118	175	237
5		9.4.6	52	5	265	80	78	134	199
5		9.4.7	83	2	263	179	84	84	263
3		9.5.1	62	6	1096	386	258	378	838
4		9.6.1							

1  
2 **Option 2d: All ecoregions**  
3 In this approach, each ecoregion (level 3) is represented by CLs that are located from sites within  
4 the spatial boundaries of the same ecoregion (level 3). Eighty-five ecoregions (level 3) occur in  
5 the conterminous U.S., 76 of those are represented by CLs in this analysis. The remaining 9  
6 ecoregions (level 3) for which CLs are not available could be represented by deposition metrics  
7 that are calculated using all of the CL data as described in option 1. This approach yields 77  
8 deposition metrics.

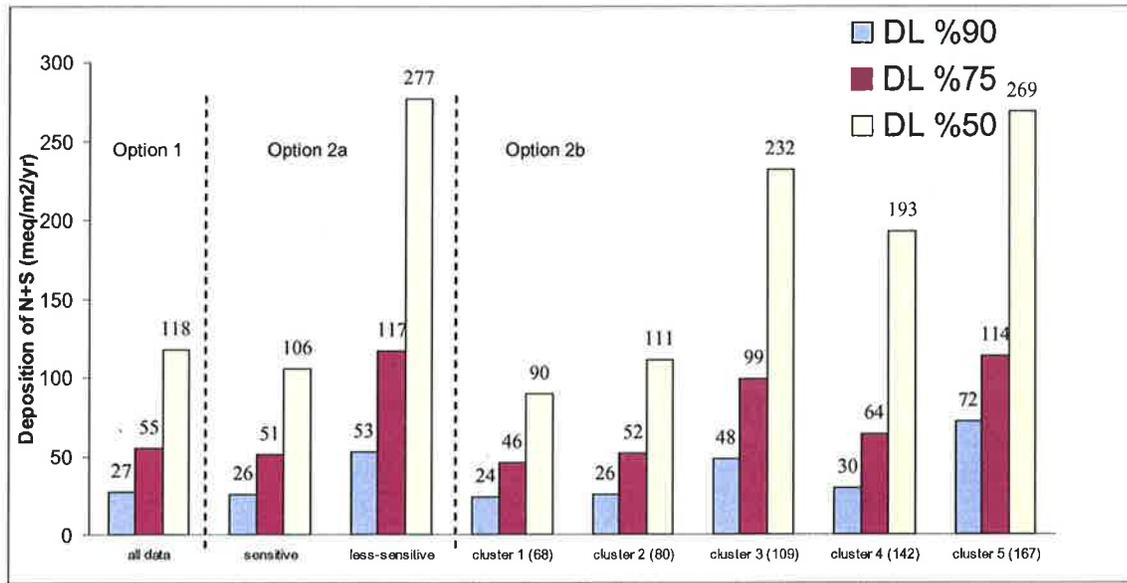
9 **Comparison**  
10 In this section, Option 1 and 2a-d are compared to help inform the decision of which  
11 option is most appropriate. Table 5.7 summarizes the number of deposition metrics that would  
12 result from each option. Option 1 has only one deposition metric. This approach would be

1 easiest to communicate and implement. However there is some concern that this approach may  
 2 under protect the most sensitive areas and over protect the least sensitive areas. Several analyses  
 3 have been made to determine if and to what extent this concern may be valid.

<b>Table 5-7 Summary of the number of deposition metrics that would result from the options for how to categorized the landscape based on acid-sensitivity</b>			
Acid-sensitivity categorization approach	#deposition metrics for less-sensitive areas	# of tradeoff curves for sensitive areas	Total N + S deposition metrics
Op#1 No subdivision of the U.S.	n/a	1	1
Op#2a Binary categorization (sensitivity based on 10 or more observations of ANC between -50 and 200 $\mu\text{eq/L}$ )	1	All sensitive areas could be represented by the same population of critical loads	2
Op#2b 5 Cluster (based on centroid of data between ANC between -50 and 20 $\mu\text{eq/L}$ )	n/a	The U.S. is categorized into 5 clusters of acid sensitivity based on ANC. Each ecoregion is assigned a cluster id, and the CLs from sites within each cluster are pooled to create the population for that cluster.	5
Op#2c 1 sensitive category and individual ecoregion categories	1	43 ecoregions (level 3) are considered sensitive, there is CL data available for 40 ecoregions at ANC= 50, the 3 ecoregions which do not have CL data could be represented by values calculated from the entire sensitive population population.	44
Op#2d All ecoregions	n/a	85 ecoregions (level 3) occur in the US, of these there is CL data for 76 therefore they could be represented by CLs only from their ecoregions. The 9 ecoregions without CLs could be represented by the entire population.	77

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1 option 1(no-subdivision/all data), 2a (sensitive vs. less-sensitive classification) and 2b (5 clusters  
 2 based on log ANC) in Fig 5.12. The deposition metric for option 1 is 27 meq/m<sup>2</sup>/yr. When  
 3 compared to the deposition metrics from option 2a, it would under protect the sensitive category  
 4 by 1 meq/m<sup>2</sup>/yr and over-protect the less-sensitive category by 27 meq/m<sup>2</sup>/yr. When option 1 is  
 5 compared to option 2b, clusters 1 and 2 would be slightly under protected by 3 and 1 meq/m<sup>2</sup>/yr,  
 6 respectively. Clusters 3, 4 and 5 would be over protected by between 3 to 35 meq/m<sup>2</sup>/yr.  
 7



8  
 9  
 10 **Figure 5-12 Deposition metrics for 90%,75% and 50% of the population for options, 1**  
 11 **(all data), 2a (binary classification), and 2b (clusters based on log ANC)**  
 12  
 13

14 A second method to compare the options 1 and 2a is presented in Fig 5.13. Sites that  
 15 would not be protected at a 90%-tile deposition metric at ANC=50  $\mu$ eq/L are shown. In both  
 16 options 10% of the total population is not going to be protected from exceeding the CL, by  
 17 comparing these maps we see that the spatial distribution of sites that are not protected between  
 18 the two options is similar. In option 2a, there are 10% of total sites that will not be protected in  
 19 both the sensitive and the less-sensitive ecoregions. This means that eventhough a region is  
 20 considered less sensitive, 10% of the water bodies would not meet the target ANC because more  
 21 deposition would be allowed. Considering the analyses represented by Fig 5.11 and 5.12, how  
 22 much benefit is gained by subdividing the landscape into acid sensitivity classes?



**One nation**

Sites for which N+S critical loads are not protected when one deposition metric to protect 90% of the population is selected. Red indicates sites not protected



**Binary categorization: sensitive and non-sensitive**

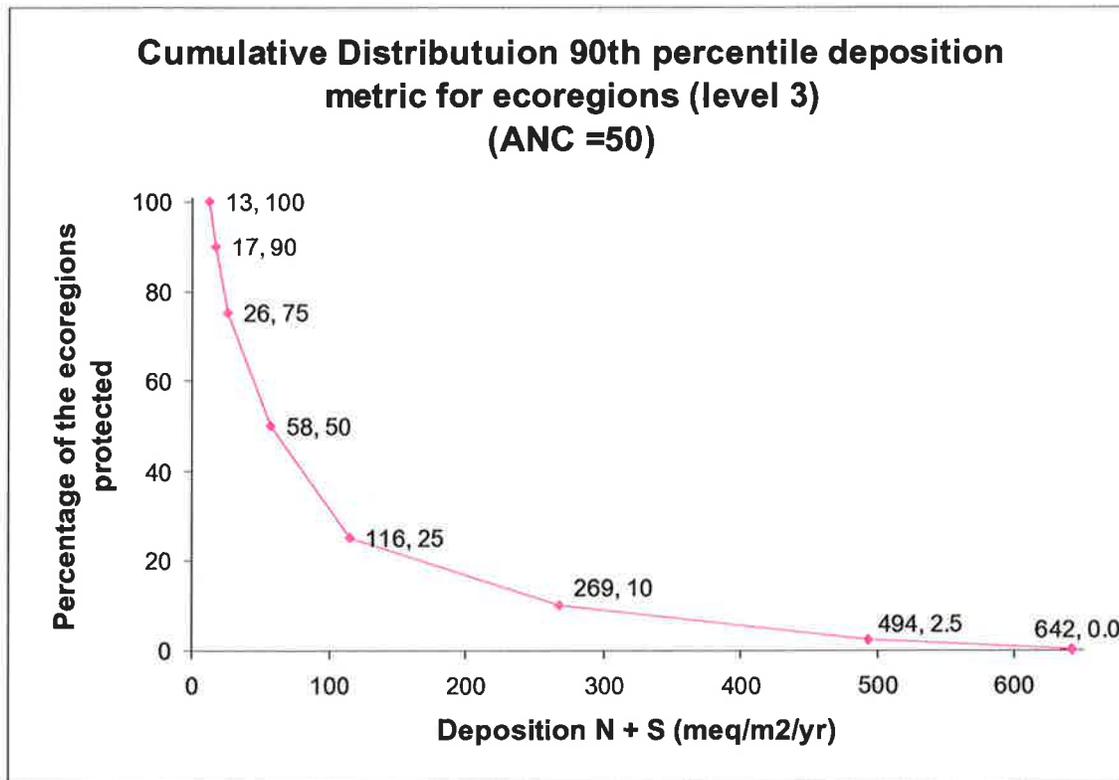
Sites for which N+S critical loads are not protected when the two deposition metrics are selected to protect 90% of each population. Red indicates sites not protected in the sensitive areas and green indicates sites that are not protected in the less-sensitive areas

1  
2 **Figure 5-13 Comparison of sites that are protected if the nation is not subdivided and if**  
3 **the nation is subdivided into a sensitive and non-sensitive category**  
4

5 Option 2c and 2d provide more spatial detail than the other options. Option 2c uses one  
6 deposition metric for all ecoregions categorized as less-sensitive areas according to option 2a,  
7 while each ecoregion considered sensitive (using log ANC) is represented only by CLs located  
8 within the boundaries of the ecoregion. Option 2d uses only CLs from within an ecoregion to  
9 represent that ecoregions. A strength of option 2d is that it provides the finest level of spatial  
10 detail consistent with available data. A weakness is that some ecoregions are represented by a  
11 small sample size of critical loads, which may not accurately convey the population of critical  
12 loads for waterbodies across the entire ecoregion. The cumulative distribution of the 90 %-tile  
13 deposition metric of the CLs at ANC=50  $\mu\text{eq/L}$  of all ecoregions (level 3) is shown in Fig 5.14.  
14 This figure helps to visualize how option 2d compares to the other options. The deposition to  
15 protect 90% of the population in the most sensitive ecoregions is 13  $\text{meq/m}^2/\text{yr}$ . In comparison  
16 to option 1, the deposition to protect 90% of the population is 29  $\text{meq/m}^2/\text{yr}$ . If the national  
17 value were chosen, then although 90% of the total population would be protected, only ~76% of

1 the ecoregions would have 90% of their populations protected. This illustrates that the sample  
2 size and distribution of critical loads varies among ecoregions.

3



4  
5

6 **Figure 5-14. Cumulative distribution of the 90% deposition metric for ecoregions is**  
7 **shown. The x and y values are given next to each data point.**

8

9 **Neco**

10 Neco is added to the deposition metric to develop the N and S tradeoff curves (See  
11 Section 5.3.2.8). Staff suggests that the spatial boundaries used to determine the acid-sensitivity  
12 categories for calculating deposition metrics are also used to calculate Neco values.

13

14 **5.3.2.8 Deposition metric: Developing N and S tradeoff curves from the deposition metrics for**  
15 **the acid sensitive categories**

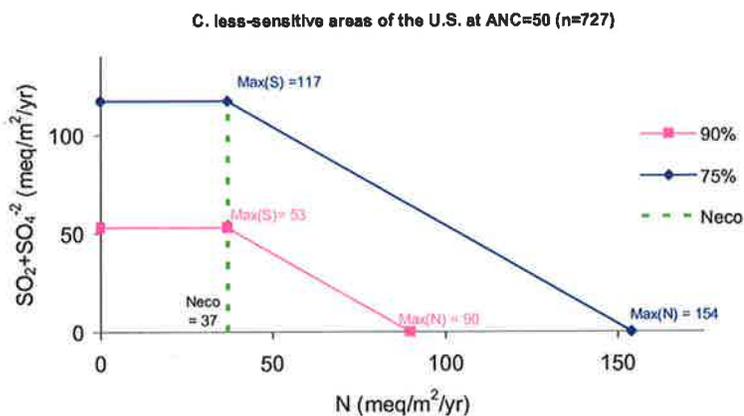
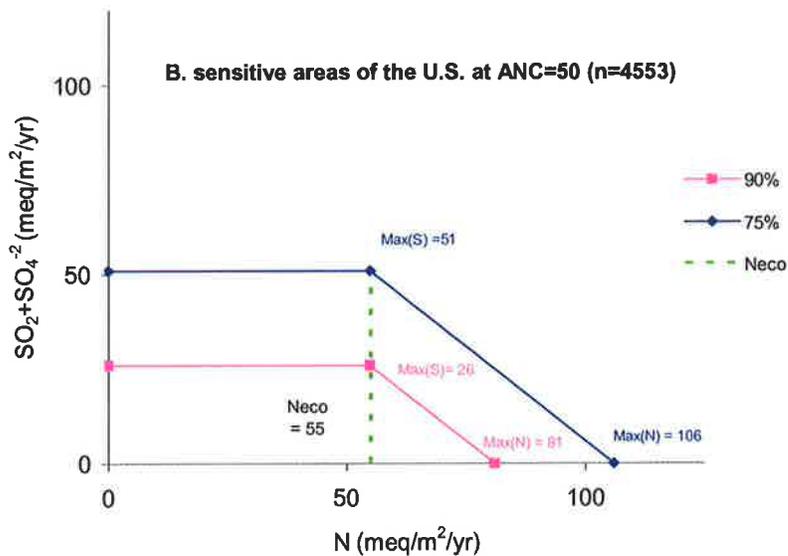
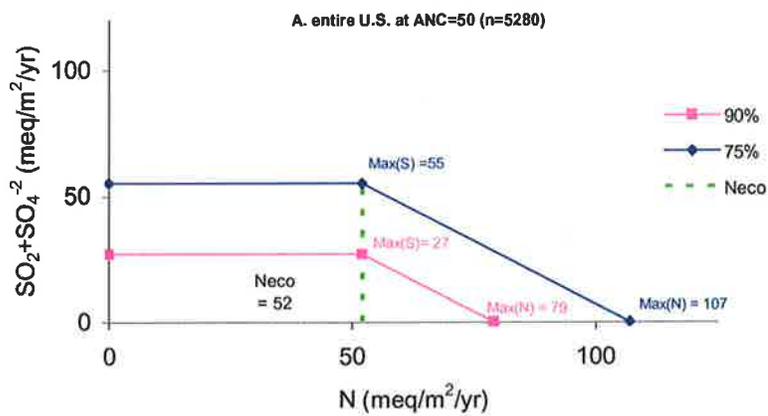
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17 Acid sensitivity categories define the spatial area of a population. Therefore the membership of  
18 an individual catchment to a population that represents an acid sensitivity category will vary  
19 depending on which approach for categorizing acid-sensitivity across the landscape is selected  
20 for the NAAQS. For each category that is established, there will be a single, specified value for  
21 a deposition metric. This deposition metric will be based on the population of catchments in that

1 specific category. Therefore both the number and the values of the deposition metrics can  
2 change based on how finely the U.S. is subdivided into acid sensitivity categories.

3         The deposition metric for a category will be a single, specified value for the deposition of  
4 N and S. That deposition load can be achieved by various combinations of N and S loading, and  
5 these combinations can be expressed as an N and S deposition tradeoff curve. Thus for each  
6 category a specific deposition load and a resulting N and S tradeoff curve can be developed.  
7 Regardless of which option is chosen for spatially dividing the U.S. into acid-sensitivity  
8 categories, the number of N and S deposition tradeoff curves is the same as the number of  
9 deposition metrics (Table 5.7). For each category, the assigned value for Neco is added to the  
10 deposition metric to develop the N and S tradeoff curve. Fig. 5.15 provides an example for two  
11 of the categorization options – Option 1, with a tradeoff curve for the single national category,  
12 reflecting the entire population of critical loads, and Option 2a, with a tradeoff curve for the  
13 sensitive and not sensitive binary landscape categorization.

14



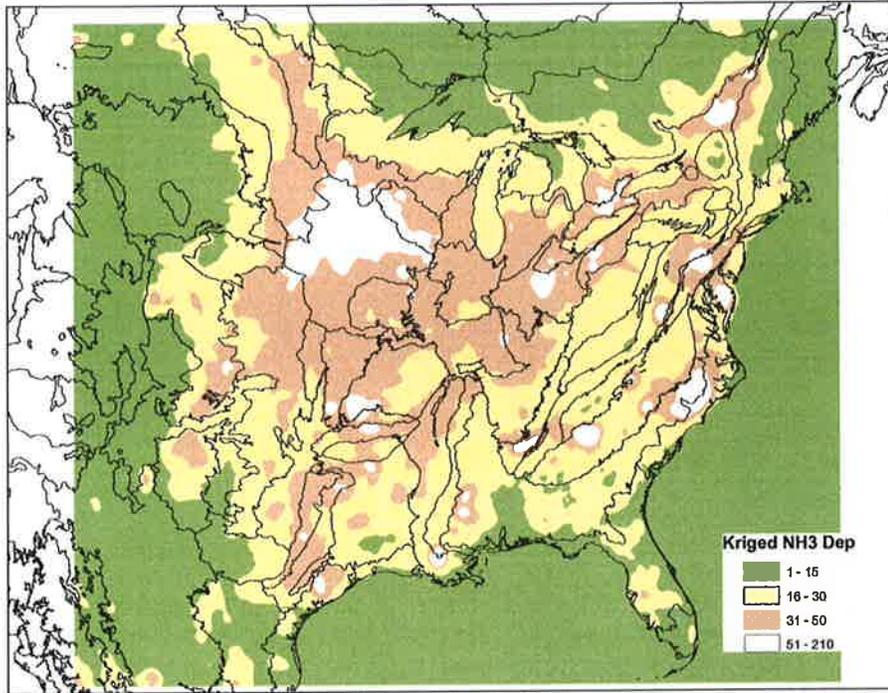
1  
2 **Figure 5-15** Examples of N + (SO<sub>2</sub> + SO<sub>4</sub><sup>-2</sup>) deposition tradeoff curves for option 1 (A) and  
3 option 2a(B and C).  
4

1  
2 **5.3.9 Deposition metric:** How is reduced nitrogen appropriately considered in the deposition  
3 metric?  
4

5 N deposition is composed of NH<sub>x</sub> deposition and NO<sub>y</sub> deposition. Deposition from NH<sub>x</sub>  
6 can contribute to acidification. However, the criteria pollutant listed by EPA pursuant to section  
7 108 (a) of the Act is oxides of nitrogen, which does not include NH<sub>x</sub>. The NAAQS is for oxides  
8 of nitrogen and sulfur, hence the form of the NAAQS needs to account for the deposition effects  
9 of these oxides. To accomplish this, the loadings of reduced forms of nitrogen derived for a  
10 given spatial area would be subtracted from the N +S deposition metric after N<sub>ECO</sub> is added, such  
11 that the resultant deposition metric is for sulfur and oxidized nitrogen only. Subtraction of  
12 reduced nitrogen from the deposition metric based on nitrogen and sulfur is expressed by  
13 equation

$$14 \quad DL_{\%ECO}(NO_Y + S) = DL_{\%ECO}(N + S) + N_{Eco} - Dep_{NHx}^{Total} \quad (6)$$

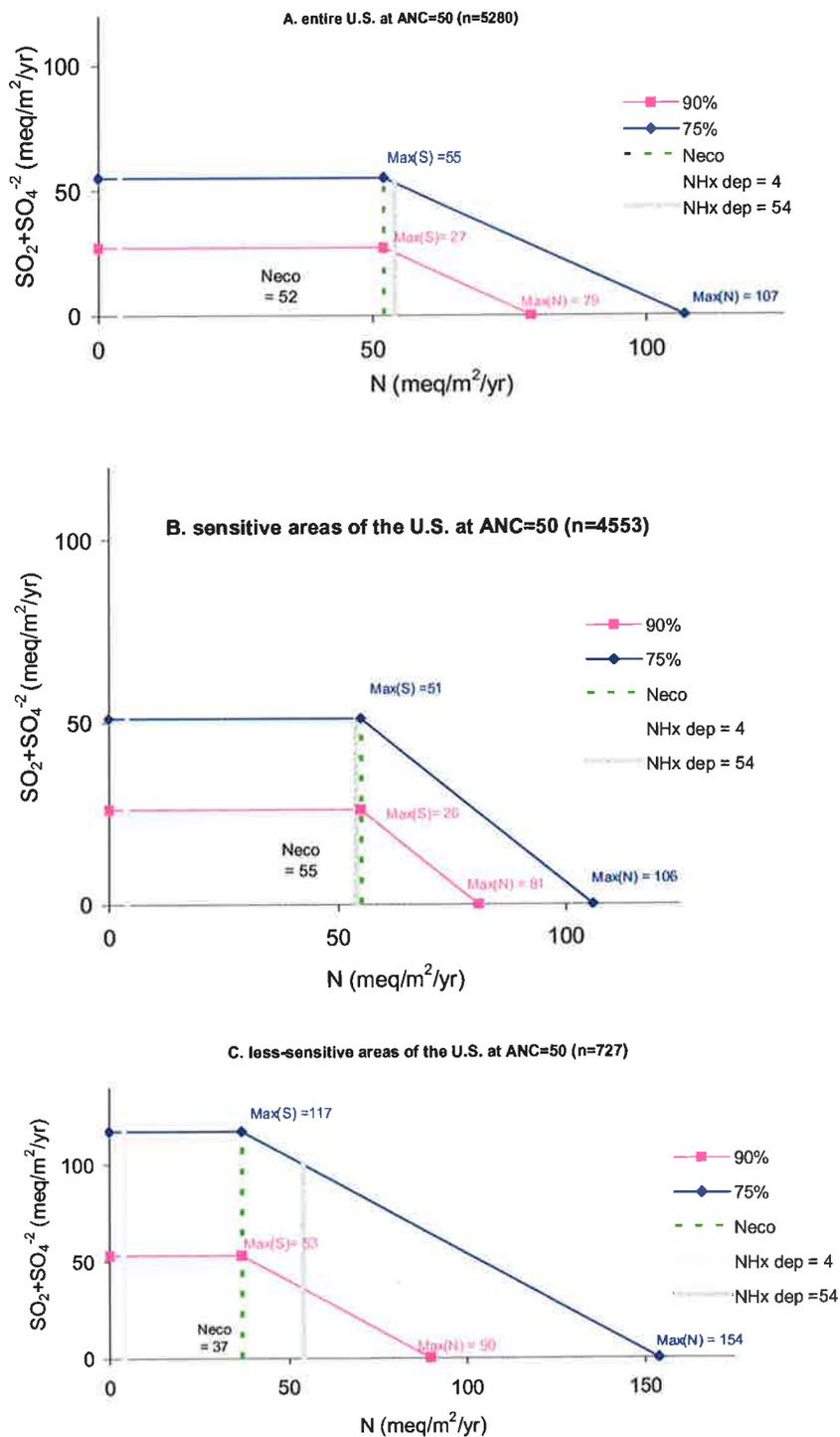
15 **Staff propose the value of reduced nitrogen is initially set using deposition of NH<sub>x</sub>**  
16 **modeled using the CMAQ, evaluated for the period 2002-2005.** The average values of NH<sub>x</sub>  
17 deposition for the 83 ecoregions (level 3) in the U.S. ranges from 4 -54 meq/m<sup>2</sup>/yr. This would  
18 mean that each acid-sensitivity category would have one N+S deposition metric, one N<sub>ECO</sub> value  
19 and one NH<sub>x</sub> deposition value. **Staff is considering the most appropriate spatial averaging**  
20 **extent for NH<sub>x</sub>. Figure 5.16 shows spatially interpolated values of reduced nitrogen**  
21 **deposition based on 12km CMAQ modeling in the Eastern U.S. It is clear that in some**  
22 **locations, there is significant heterogeneity in NH<sub>x</sub> deposition within ecoregion 3**  
23 **boundaries. Given this information, two possible approaches to estimating reduced**  
24 **nitrogen values are possible: 1) average reduced nitrogen deposition within an ecoregion,**  
25 **acknowledging that this will lead to uncertainties in the level of protection associated with**  
26 **levels of ambient NO<sub>y</sub> and SO<sub>x</sub>, or 2) allow for additional spatial refinement of sensitive**  
27 **areas to reflect the heterogeneity of NH<sub>x</sub> deposition.**



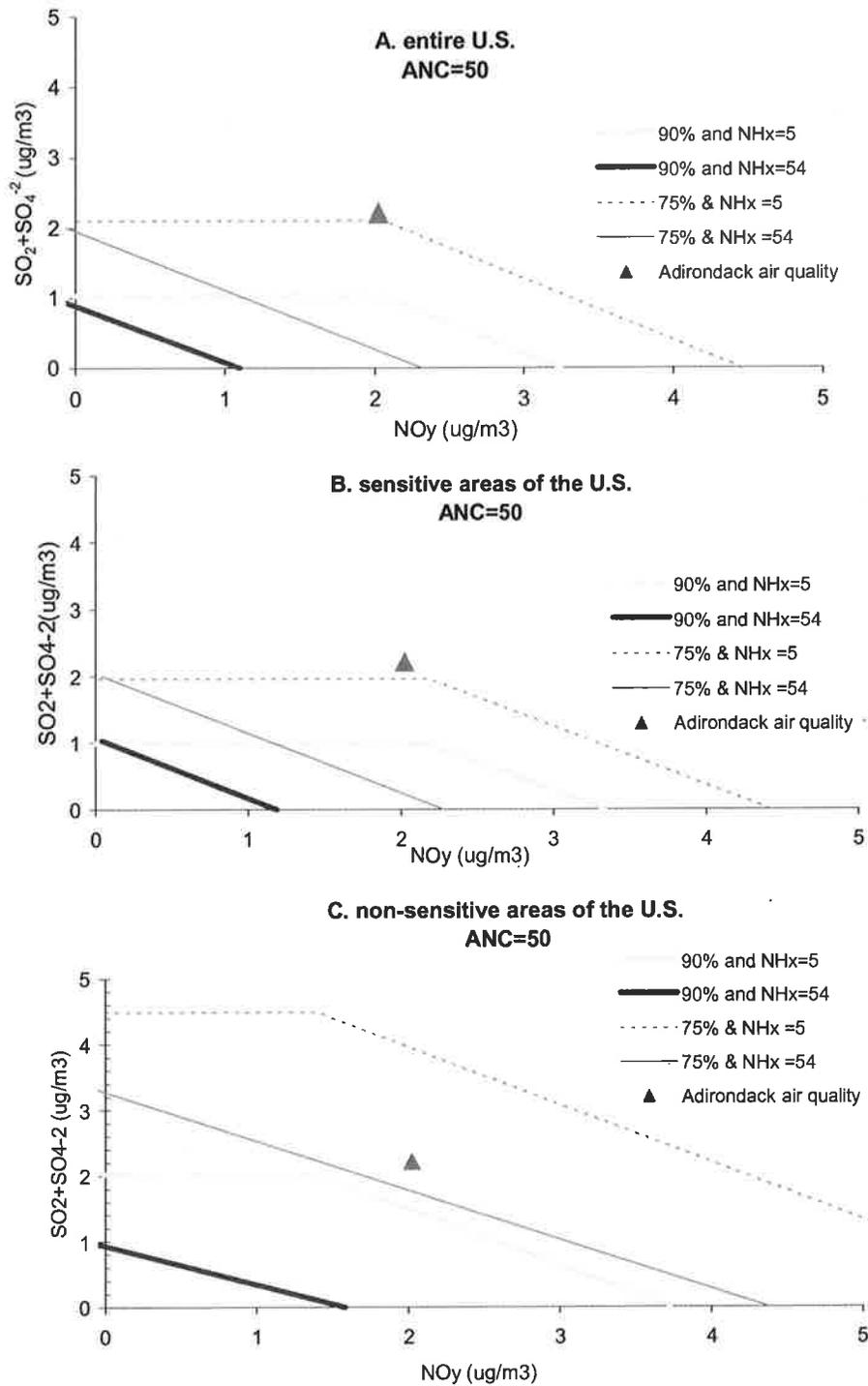
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**Figure 5-16. A U.S. map of NH<sub>3</sub> deposition (meq/m<sup>2</sup>/yr) overlaying ecoregions (level 3) boundaries**

The deposition from NO<sub>y</sub> and SO<sub>x</sub> is converted to atmospheric concentrations of NO<sub>y</sub> and SO<sub>x</sub> by the methods described in section 5.2.3 and an example is given in fig 5.19. This figure also shows air quality in the Adirondack region relative to the NO<sub>y</sub> and SO<sub>x</sub> tradeoff curves for option 1 and option 2a. In option 1 the Adirondacks air quality is slightly out of attainment for a 75%-tile deposition metric based on CL at ANC=50. In option 2a the Adirondack air quality is out of attainment for the curve for the sensitive areas, but in attainment for the less-sensitive areas.



1  
 2 **Figure 5-18. Examples of N + (SO<sub>2</sub> + SO<sub>4</sub><sup>-2</sup>) deposition tradeoff curves for option 1**  
 3 **(A) and option 2a(B and C). Deposition metrics that correspond to protection 75% or 90%**  
 4 **of ecosystems are given with the high and low value for average NHx deposition in**  
**ecoregions indicated by vertical grey lines.**



**Figure 5-19.** Examples of NOy + (SO<sub>2</sub> + SO<sub>4</sub><sup>-2</sup>) deposition tradeoff curves for option 1 (A) and option 2a(B and C). Deposition metrics that correspond to protection 75% or 90% of the ecosystems considering both the high and low value for NHx deposition

### 5.3.3 Conceptual Design: Linking Deposition to Atmospheric Concentration

#### 5.3.3.1 Background

Atmospheric pollutants deposit onto land and water surfaces through at least two major mechanisms: direct contact with the surface (dry deposition), and transfer into liquid precipitation (wet deposition). A third mechanism involving impaction of fog droplets onto the surface (occult deposition) also exists, but has been shown to be very small relative to wet deposition (Fuhrer 1986; Thalmann 2001) and so is not considered in calculations below. The magnitude of each deposition process is related to the ambient concentration through the time-, location-, process- and chemical species-specific *deposition velocity* (Seinfeld and Pandis, 1998):

$$Dep_i^{Dry} = v_i^{Dry} \cdot C_i^{Amb} \quad (7)$$

$$Dep_i^{Wet} = v_i^{Wet} \cdot C_i^{Amb} \quad (8)$$

where  $v_i^{Dry}$  and  $v_i^{Wet}$  are the dry and wet deposition velocities,  $Dep_i^{Dry}$  and  $Dep_i^{Wet}$  are the dry and wet deposition fluxes,  $C_i^{Amb}$  is the ambient concentration, and the  $i$  subscript indicates the pollutant species under study. The total deposition of each pollutant is

$$Dep_i^{Tot} = Dep_i^{Dry} + Dep_i^{Wet} \quad (9)$$

Substituting Equations 7 and 8 into Equation 9 yields

$$Dep_i^{Tot} = v_i^{Dry} \cdot C_i^{Amb} + v_i^{Wet} \cdot C_i^{Amb} \quad (10)$$

The total deposition of sulfur or nitrogen would therefore be:

$$Dep_\alpha^{Tot} = \sum_i (v_i^{Dry} + v_i^{Wet}) \cdot m_i \cdot C_i^{Amb} \quad (11)$$

where  $m_i$  is the molar ratio of the atom of interest ( $\alpha$ , which refers to either sulfur or nitrogen atoms in  $SO_x$  or  $NO_y$ ) to the  $i$ 'th pollutant. Note that only species in  $SO_x$  and  $NO_y$  (see Table 1) will be input to these equations;  $NH_3$  and  $NH_4$  are not currently included as listed pollutants (see Chapter 8 for an expanded discussion of the role of  $NH_x$ ).

#### 5.3.3.2 Aggregation Issues

A relationship for converting sulfur or nitrogen deposition to “equivalent” ambient concentrations, which is part of specifying ambient air quality standards, is

1 provided in equation 11. A major issue to consider during such conversion is the  
2 treatment of spatial, temporal and chemical resolutions of the deposition data and the  
3 resulting standards. Since the objective is to specify allowable levels of total ambient  
4 SO<sub>x</sub> and NO<sub>y</sub> (as S and N),, and this is also the chemical resolution provided by the  
5 ecosystem models, it is convenient to use a relationship with the following form:

$$6 \quad Dep_i^{Tot} = T_i \cdot C_i^{Amb} \quad (12)$$

7 where  $T_i$  is what we term the “transference ratio”, which can be considered an  
8 aggregated, “effective” deposition velocity that relates total deposition of sulfur or  
9 nitrogen to the total ambient concentration, and represents an average of the chemical  
10 species specific  $v_i^{Tot}$  ( $= v_i^{Dry} + v_i^{Wet}$ ) values in Equation 11. In other words, the  
11 transference ratio is effectively the aggregate deposition velocity that would have been  
12 required to obtain a level of deposition given a level of atmospheric concentrations. The  
13 SO<sub>x</sub> and NO<sub>y</sub> concentrations are the result of applying the  $m_i$  values to the  $C_i^{Amb}$  values  
14 in Equation 11. Although  $T_i$  values are representative of pure nitrogen and sulfur, they  
15 are derived from concentrations and depositions of the individual pollutants, so that the  
16 resulting value is appropriately weighted by the relative levels of each pollutant actually  
17 present in the environment.

18 The deposition velocities (and, by extension, the  $T_i$  values) provide relationships  
19 between depositions and concentrations that are concurrent. Since the deposition critical  
20 loads derived from ecosystem models that are in need of conversion to atmospheric  
21 concentrations are in terms of annual deposition, the ambient concentrations will also be  
22 aggregated to the annual level through averaging. Data used to derive annual  $T_i$  values  
23 will also need to have the same spatial representativeness as the depositional loads  
24 derived from ecosystem models. However, many of the calculations involving  
25 relationships between concentrations and depositions will be in terms of the masses of the  
26 N and S atoms within NO<sub>y</sub> and SO<sub>x</sub> removed from the other atoms in the pollutant  
27 species, which we will refer to as “oxidized S” and “oxidized N”. This is done both  
28 because these quantities are the outputs of the ecosystem models discussed in Section  
29 XXX, and it removes potential ambiguities that might be introduced by different species  
30 mixtures.

31

### 1 5.3.3.3 Air Quality Simulation Models

2 Ideally,  $T_i$  values would be derived for each area of interest from concurrently  
3 collected sulfur and nitrogen deposition and concentration measurements. However, no  
4 monitoring network currently exists that can provide such information. We therefore  
5 suggest using output of the Community Multi-scale Air Quality (CMAQ) model (EPA,  
6 1999) for initial calculation of  $T_i$  values.

7 CMAQ provides both concentrations and depositions for a large suite of pollutant  
8 species on an hourly basis for 12 km grids across the continental U.S. Its comprehensive  
9 structure is ideal for providing  $T_i$  values that appropriately address the chemical and  
10 temporal aggregation issues discussed above, and weighted spatial averages of the  
11 gridded data can be used for areas that span multiple grid cells. The major potential  
12 drawback to using CMAQ output is that the data is simulated rather than measured.

13 CMAQ does not directly calculate or use  $T_i$  values; instead the following  
14 procedures are used in the code to model deposition:

15 1)  $v^{dry}$  values of gaseous pollutants are calculated in the CMAQ weather module  
16 called the Meteorology-Chemistry Interface Processor (MCIP) through a complex  
17 function of meteorological parameters (e.g. temperature, relative humidity) and properties  
18 of the geographic surface (e.g. leaf area index, surface wetness)

19 2)  $v^{dry}$  values for particulate pollutants are calculated in the aerosol module of  
20 CMAQ, which, in addition to the parameters needed for the gaseous calculations, also  
21 accounts for properties of the aerosol size distribution

22 3)  $v^{wet}$  values are not explicitly calculated. Wet deposition is derived from the  
23 cloud processing module of CMAQ, which performs simulations of mass transfer into  
24 cloud droplets and aqueous chemistry to incorporate pollutants into rainwater, all of  
25 which is conceptually contained in the  $v^{wet}$  parameter in Equation 8.

26 Due to lack of direct measurements, no performance evaluations of CMAQ's dry  
27 deposition calculations can be found; however, the current state of MCIP is the product  
28 of research that has been based on peer-reviewed literature from the past two decades  
29 (EPA, 1999) and is considered to be EPA's best estimate of dry deposition velocities.  
30 Some bias has been found between CMAQ's wet deposition predictions and measured  
31 values (Morris et al., 2005); recent analyses suggest that poor simulation of precipitation

1 could be responsible for this (Davis and Swall, 2006), which can potentially be dealt with  
2 by recalculating wet deposition using precipitation measurements. Although the model is  
3 continually undergoing improvement, CMAQ is EPA's state-of-the-science  
4 computational framework for calculating deposition velocities, and was therefore the  
5 logical first choice as a source for  $T_i$  values.

#### 6 7 **5.3.3.4 Oxidized Sulfur and Nitrogen Pollutant Species**

8 Ideally, all possible NO<sub>y</sub> and SO<sub>x</sub> air pollutant species that contribute to  
9 ecological adversity would be considered for  $T_i$  values. The pollutant list is constrained  
10 by the source of  $T_i$  values, which is currently CMAQ output. The oxidized sulfur and  
11 nitrogen species currently available in CMAQ whose data will be used for  $T_i$  values, are  
12 listed in Table 1.

13 One issue that needs explicit consideration is the contributions of particles larger  
14 than PM<sub>2.5</sub> to sulfur and nitrogen deposition. A recent review of particle deposition  
15 measurements (Grantz, Garner, and Johnson, 2003) showed that coarse particles  
16 generally deposit far more sulfate and nitrate in forest ecosystems than fine particles.  
17 However, CMAQ does not currently provide simulations of coarse particulate sulfate and  
18 nitrate. This is an issue that needs to be addressed by developers of either the model or  
19 the future SO<sub>x</sub>/NO<sub>y</sub> measurement network to set scientifically sound standards. This  
20 issue is explored further in the discussion of monitoring in Section XXX.

#### 21 **5.4.5 Example Calculations**

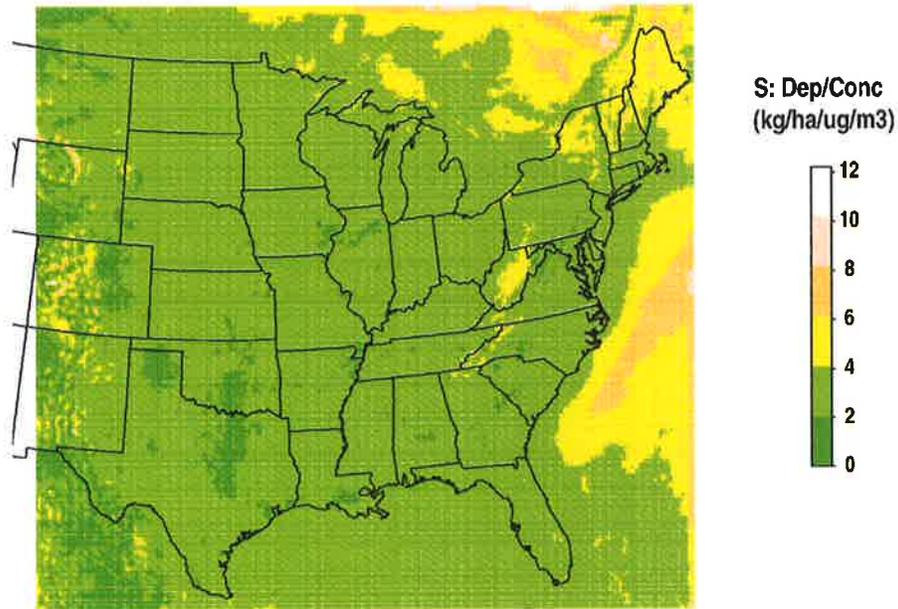
22 Figure 5.20 shows annual inverse  $T_i$  values<sup>1</sup> calculated for each 12 km grid in the  
23 eastern and western domains for a 2002 CMAQ v4.6 simulation, which is the quantity  
24 that would be used for conversion of deposition load tradeoff curves. Figure 5.21 shows  
25 an example application of these ratios for a lake in the Adirondacks. Deposition load  
26 tradeoff curves for this lake (see Section 5.5 for their calculation) are multiplied by the  
27 inverse  $T_i$  value from the appropriate gridcell in Fig. 5.22 to convert those depositions to  
28 ambient concentrations of sulfur and nitrogen.

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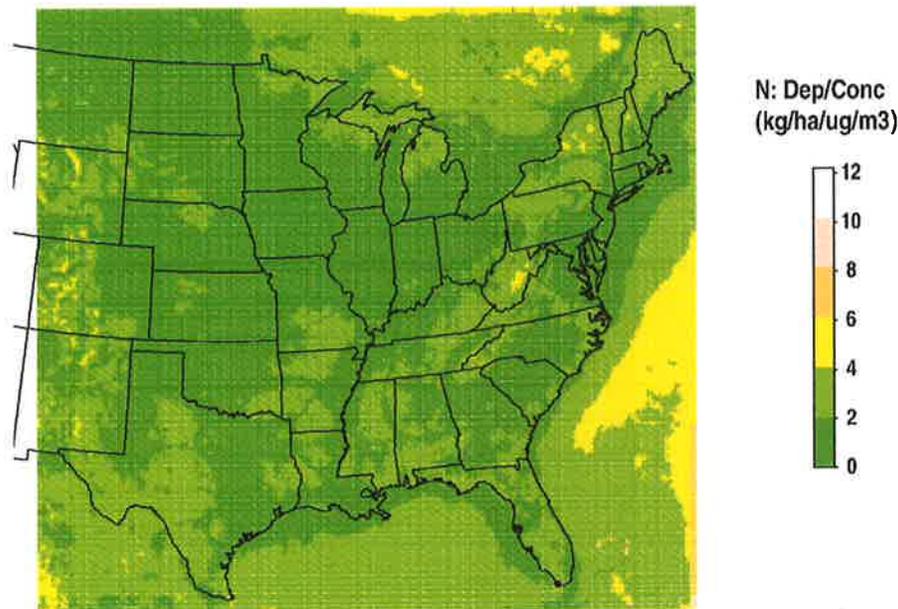
<sup>1</sup> Inverse  $T_i$  values represent the multiplier needed to convert deposition levels into atmospheric concentrations of NO<sub>y</sub> and SO<sub>x</sub>.

1           A CMAQ v4.7 simulation for multiple years (2002-2005) recently became  
2 available, which was used to examine the inter-annual variability of inverse  $T_i$  values.  
3 The grid-specific coefficients of variation (CV) are shown in Fig. 5.22. Fig. 5.22 shows  
4 that CV values are relatively small (< 25%) in the Adirondacks and Shenandoah case  
5 study areas. This suggests that a 3-year average of the ratios may be a sufficiently stable  
6 representation of deposition velocities for converting the deposition load curves to  
7 ambient concentrations.

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11  
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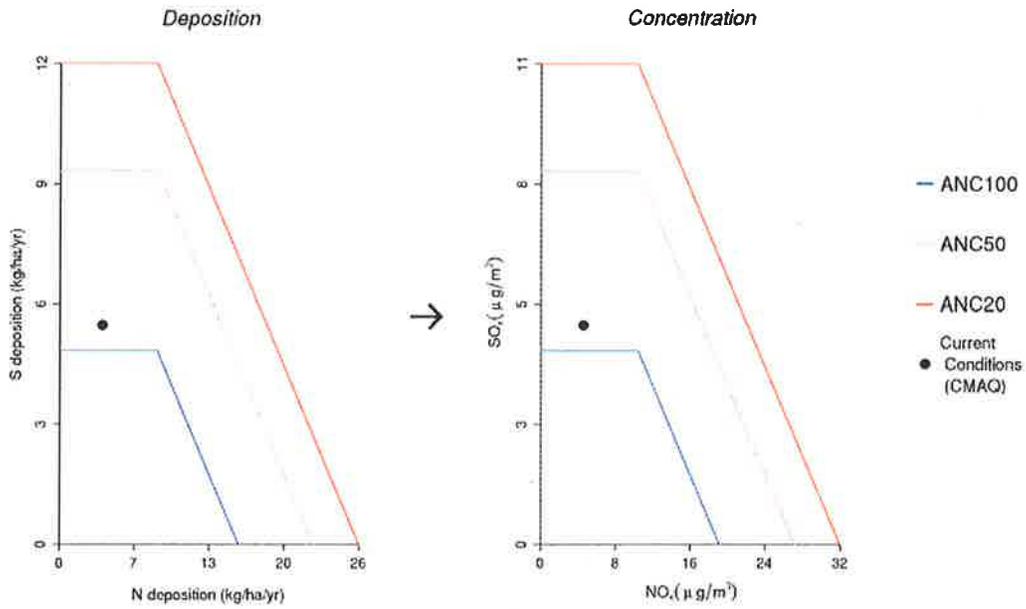


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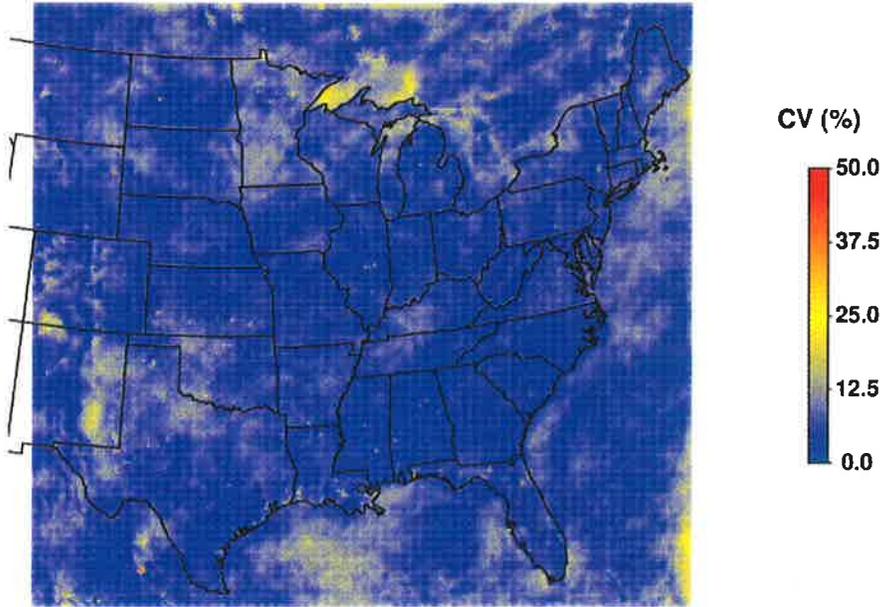
**Figure 5-20 2005  $T_i$  values for each grid cell in the eastern U.S. domain.** The top map shows values for sulfur and the bottom is for nitrogen.



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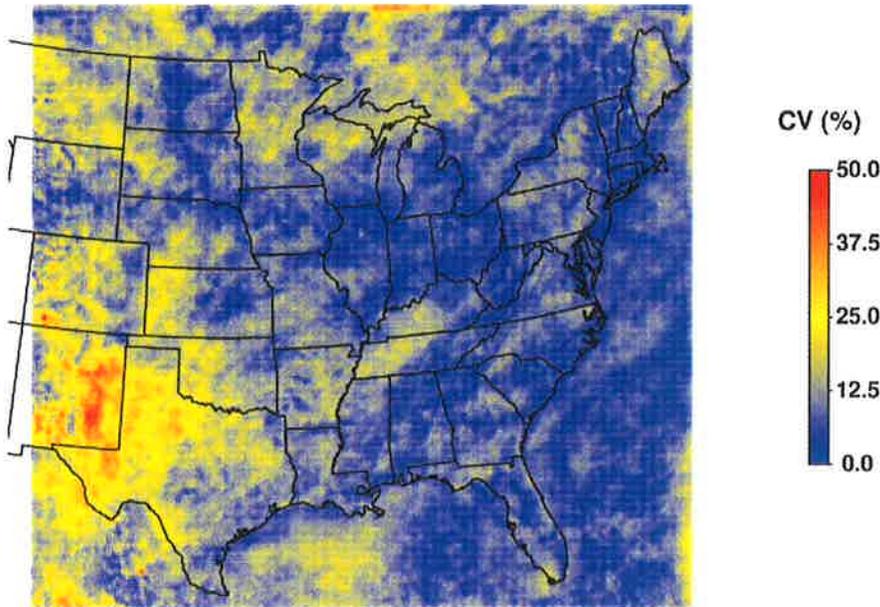
**Figure 5-21. Schematic Diagram illustrating the procedure for converting deposition tradeoff curves of sulfur and nitrogen to atmospheric concentrations of SO<sub>x</sub> and NO<sub>y</sub>.**

Coefficient of Variation of N Conc/Dep ratio, 2002-2005



1 a)

Coefficient of Variation of S Conc/Dep ratio, 2002-2005



2 b)

3

4

5

6

**Figure 5-22 Inter-annual coefficients of variation (CV) of a)  $\text{NO}_y$  and b)  $\text{SO}_x T_i$  values, based on a series of 2002-2005 CMAQ v4.7 simulation.**

1 **5.3.3 AAPI**

2  
3 In sections 5.3.2 and 5.3.3, the form of the standard is expressed as an approach for  
4 linking ecosystems, N and S deposition and ambient air concentrations of NOx and SOx  
5 conceptually identify levels of NOx and SOx that are protective of ecosystems via NOx and SOx  
6 tradeoff curves. The tradeoff curves will differ among acid-sensitivity categories because they  
7 are derived from a specific percentage of ecosystems protected within an overall sensitive area.  
8 In this section, the form of the standard is expressed as the AAPI equation, which is an index, set  
9 to one level, which can be applied across the nation to convey the potential of an ecosystem to  
10 become acidified from atmospheric deposition. Moreover, the AAPI calculates the target ANC  
11 for a percentage of aquatic ecosystems within a particular acid sensitive area when atmospheric  
12 concentrations of NOx and SOx are input  
13

14 **5.3.4.2 Definition and Derivation of the AAPI**

15 The definition of the AAPI considered here is:

16  
17 Annual Average AAPI: Natural background ANC minus the contribution to acidifying  
18 deposition from NHx, minus the acidifying contribution of deposition from NOy and  
19 SOx.  
20

21 In order to derive the AAPI, we start with the basic framework of critical loads discussed in  
22 section 5.3. The approach used to calculate N and S deposition values for a specified ANC at a  
23 catchment-scale is expressed in Equation 2 from section 5.3.  
24

25 
$$CL_{ANC\lim}(N + S) = ([BC]_O^* - [ANC_{\lim}])Q + N_{eco} \quad (2)$$

26  
27 To develop an equation that allows us to set a single value for the standard across the U.S.,  
28 rearrange equation (2) to solve for ANC (place ANC on the left hand side of the equation):  
29

30 
$$Q \cdot ANC_{\lim} = N_{ECO} + [BC]_O^* \cdot Q - [Dep_N^{Total} + Dep_S^{Total}] \quad (13)$$

$$ANC_{lim} = \frac{1}{Q} N_{ECO} + [BC]_o^* - \frac{1}{Q} [Dep_N^{Total} + Dep_S^{Total}] \quad (14)$$

Building from equation 14, total nitrogen deposition is split into oxidized and reduced nitrogen because we need to be able to specify the standards in terms of oxides of nitrogen, and so the contribution of reduced nitrogen has to be separated.

$$ANC_{lim} = \left[ \frac{1}{Q} \cdot N_{ECO} + [BC]_o^* \right] - \frac{1}{Q} [Dep_{NOy}^{Total} + Dep_S^{Total}] - \frac{1}{Q} \cdot Dep_{NHx}^{Total} \quad (15)$$

Where,

$Dep_{NHx}^{Total}$  = the depositional load of reduced nitrogen, NHx.

In order to judge whether a catchment meets the  $ANC_{limit}$  given observed NOy and SOx levels, the associated depositional loadings of NOy and S can be compared directly against calculated deposition tradeoff curves, atmospheric concentrations of NOy and SOx can be compared against the atmospheric concentration tradeoff curves or, loadings of NOy and SOx can be input into the following equations to obtain the calculated value of ANC, equal to  $ANC^*$ :

$$ANC^* = \left[ \frac{1}{Q} \cdot N_{ECO} + [BC]_o^* \right] - [L(NOy) + L(SOx)] - L(NHx) \quad (16)$$

Where,

$ANC^*$  = the calculated value of ANC given loadings of N and S for comparison against an  $ANC_{limit}$ .

$L(NOy+S)$  = the load of NOy+S anions based on observed atmospheric concentrations of NOy and SOx

$L(NHx)$  = the load of reduced nitrogen deposition

[Note that  $L(N) = L(NOy+NHx)$ ]

1 In equation 16, the ANC\* will vary based on the deposition load inputs of NOx, NHx and  
 2 S at the site of interest. The deposition loads caused by NOx and S and NHx are inputs, leading  
 3 to

$$4 \quad ANC^* = \left[ \frac{1}{Q} \cdot N_{ECO} + [BC]_0^* \right] - \frac{1}{Q} [Dep_{NOy}^{Total} + Dep_S^{Total}] - \frac{1}{Q} \cdot Dep_{NHx}^{Total} \quad (17)$$

6 If  $ANC^* < ANC_{lim}$ , then the deposition of N and S exceeds the deposition load to maintain  
 7  $ANC_{limit}$ .  $ANC^*$  is still representative of the calculated ANC based on specific catchment level  
 8 estimates of  $[BC]_0^*$ ,  $N_{ECO}$ ,  $Q$  and  $NH_x$ .

9 The AAPI is equivalent to the equation for calculating  $ANC^*$  when the catchment  
 10 specific values are replaced by spatially aggregated values that represent an acid sensitive areas  
 11 (based on a percentile of water bodies targeted for an ANC level selected by the Administrator),  
 12  $NH_x$  is replaced by average values for aggregate ecosystem areas, and aggregated deposition  
 13 velocities translate NOy and SOx into deposition:

$$15 \quad AAPI = \left[ \frac{1}{Q} \cdot N_{eco} + [BC]_{\%eco}^* \right] - \frac{1}{Q} Dep_{\alpha NHx}^{Total} - \frac{1}{Q} [T_{NOy} \cdot C_{NOy}^{amb} + T_{SOx} \cdot C_{SOx}^{amb}] \quad (18)$$

17 Where  $C_{NOy}^{amb}$  and  $C_{SOx}^{amb}$  are concentrations of NOy and SOx, respectively,  $T_{NOy}$  and  $T_{SOx}$  are the  
 18 transfer ratios to convert ambient concentrations to deposition for NOy and SOx, respectively  
 19 (See section 5.3.3 for further description of calculation of these ratios)

20 Note that while equation (18) is used to calculate the value of AAPI for any observed  
 21 values of NOx and SOx, the level of the standard for AAPI selected by the administrator should  
 22 reflect a wide number of factors, including desired level of protection indicated by a target  
 23  $ANC_{limits}$ , the specified percentile of waterbodies projected to achieve the target ANC, and the  
 24 various factors and uncertainties involved in specifying all of the other aspects of the standard,  
 25 such as the classification of landscape areas, the specification of reduced nitrogen deposition, the  
 26 methodology to determine deposition of NOx and SOx, and the averaging time. As such the  
 27 administrator may choose an AAPI level higher or lower than the target  $ANC_{limit}$  to reflect the  
 28 combined effect of the all of the components of the standard and their related uncertainty, such

1 that the chosen AAPI, in the context of the overall standard, reflects her informed judgment as to  
2 a standard that is sufficient but not more than necessary to protect against adverse public welfare  
3 effects.

#### 5 **5.3.4.2 How are AAPI parameters determined?**

6 Other than ambient levels of NO<sub>x</sub> and SO<sub>x</sub>, which would be measured values, EPA  
7 would determine and specify all of the values for the AAPI parameters, as discussed below.

8  $T_{\alpha NO_y}$  and  $T_{\alpha SO_x}$  are calculated from CMAQ by dividing the annual average NO<sub>y</sub>, SO<sub>x</sub>  
9 concentration by the total NO<sub>y</sub> or SO<sub>x</sub> deposition, respectively, for each grid cell and then  
10 aggregating all grid cells in a level three ecoregion. The  $T_{\alpha NO_y}$  and  $T_{\alpha SO_x}$  are spatially variable,  
11 and for the purposes of setting the standard, are determined based on the ratios of total sulfur and  
12 nitrogen depositions to concentrations from CMAQ model outputs (as described in section 5.3).

13 NH<sub>x</sub> is spatially variable and determined based on monitored and/or CMAQ modeled  
14 outputs. The average NH<sub>x</sub> deposition across grid cells within an acid sensitive region will be  
15 used to represent the depositional load of NH<sub>x</sub>. (See section 5.3)

16 There will be multiple combinations of concentrations of NO<sub>y</sub> and SO<sub>x</sub> that result in a  
17 specific value of the AAPI. There will be no single combination of NO<sub>y</sub> and SO<sub>x</sub> that solves for  
18 a particular value of AAPI in all locations. Measured concentrations of annual average NO<sub>y</sub> and  
19 SO<sub>x</sub> necessary to meet the standards are thus expressed conditionally by the equality in the  
20 AAPI equation (18), and not by fixed quantities.

21 In order to provide a set of values for elements of the form, the  $T_{NO_y}$ ,  $T_{SO_x}$  and NH<sub>x</sub>,  
22 values for specific areas would be estimated based on the best available monitoring and/or  
23 modeling data. Given the limited availability of measured deposition velocities, staff concludes  
24 that the calculated deposition ratios based on the CMAQ modeling from 2005 provides the best  
25 available source of estimates of  $T_{NO_y}$  and  $T_{SO_x}$ . Evaluation of the stability of these estimates of  
26 deposition ratios over time suggests that in most acid sensitive areas, deposition ratios are quite  
27 stable, with a coefficient of variation less than 25 percent across a four year period. While there  
28 are a limited number of sites that directly measure deposition of reduced nitrogen, staff

1 concludes that the most widely available and defensible estimates of reduced nitrogen deposition  
 2 (NHx) are the estimates obtained from the CMAQ modeling from 2005.<sup>2</sup>

3 For each acid sensitive area the natural background ANC is a calculated value and is  
 4 determined by the expression  $\left[ \frac{1}{Q} \cdot N_{eco} + [BC]_O^* \right]_{\%eco}$  from equation 18. The three components  
 5 are:  $[BC]_O^*$ , the pre-industrial weathering rate;  $N_{eco}$ , which represents the amount of deposited  
 6 nitrogen that is removed via ecosystem uptake, denitrification and immobilization; and  $Q$ , the  
 7 runoff parameter. These components act in combination, so that the 95<sup>th</sup> percentile of the  
 8 distribution of values for each variable may not equal the 95<sup>th</sup> percentile value for the  
 9 combination of the variables. Therefore the value for the combination of these variables will be  
 10 determine from the critical load which represents the selected % of protection given the  
 11 distribution of critical loads in the population.

12  
 13 It is important to note for this form of the standard that the same AAPI can be obtained  
 14 with different combinations of ambient NOy and SOx concentrations. The implication of the  
 15 form of the standard expressed in equation (18) is that there will be a tradeoff curve that reflects  
 16 the combinations of NOy and SOx that satisfy equation (18) for any specific value of the  
 17 standard. The shape of the tradeoff curve will depend on the specific values of

18  $\left[ \frac{1}{Q} \cdot N_{eco} + [BC]_O^* \right]_{\%eco}$ ,  $T_{NOy}$ ,  $T_{SOx}$  and NHx for a limited number of specific areas classified

19 based on acid-sensitivity. As discussed in Section 5.3, all parts of the U.S. would be classified  
 20 into areas based on acid-sensitivity. Within each such area, EPA would specify the parameter  
 21 values of AAPI, leading to a specific tradeoff curve for each area.

22 The levels of NOy and SOx that meet an AAPI standard expressed for a given

23  $\left[ \frac{1}{Q} \cdot N_{eco} + [BC]_O^* \right]_{\%eco}$ ,  $T_{NOy}$ ,  $T_{SOx}$  and NHx:

24  
 25 
$$\left[ T_{NOy} \cdot C_{NOy}^{amb} + T_{SOx} \cdot C_{SOx}^{amb} \right] = Q \left[ \frac{1}{Q} \cdot N_{eco} + [BC]_O^* \right]_{\%eco} - Dep_{NHx}^{Total} - Q \cdot AAPI \quad (19)$$

26

---

<sup>2</sup> Note to readers: Maps of CMAQ 2005 estimates of NHx deposition will be included in the second draft policy assessment, along with an evaluation of the representativeness of the 2005 NHx deposition for characterizing conditions over a multiyear period.

1 Note that  $[T_{NO_y} \cdot C_{NO_y}^{amb} + T_{SO_x} \cdot C_{SO_x}^{amb}]$  is essentially the critical load of NO<sub>y</sub> and S expressed in  
 2 terms of atmospheric concentrations. The pairs of NO<sub>y</sub> and SO<sub>x</sub> that will meet a given AAPI  
 3 limit are related through the following equations

$$4 \quad NO_y^* = C_{\min}(NO_y) \quad (20)$$

$$5 \quad SO_x^* = C_{\max}(SO_x) \forall NO_y < C_{\min}(NO_y) \quad (21)$$

$$6 \quad SO_x^* = C_{\max}(SO_x) + \left[ \frac{C_{\max}(SO_x)}{(C_{\min}(NO_y) - C_{\max}(NO_y))} \right] \cdot NO_y^* \forall NO_y > C_{\min}(NO_y) \quad (22)$$

8 Where,

9  $NO_y^*$  is the coordinate point for NO<sub>y</sub>

10  $SO_x^*$  is the coordinate point for SO<sub>x</sub>

11  $C_{\max}(SO_x)$  is the concentration of SO<sub>x</sub> in the atmosphere consistent with DL<sub>max</sub> (S)

12  $C_{\max}(NO_y)$  is the concentration of NO<sub>y</sub> in the atmosphere consistent with DL<sub>max</sub> (N)

13  $C_{\min}(NO_y)$  is the concentration of NO<sub>y</sub> in the atmosphere consistent with DL<sub>min</sub> (N)

14

$$15 \quad C_{\max}(SO_x) = \frac{1}{T_{aS}} DL_{\max}(S) \quad (23)$$

$$16 \quad C_{\min}(NO_y) = \frac{1}{T_{\alpha NO_y}} DL_{\min}(N - NH_x) \forall NH_x < DL_{\min}(N) \quad (24)$$

$$17 \quad = 0 \forall NH_x > DL_{\min}(N)$$

$$18 \quad C_{\max}(NO_y) = \frac{1}{T_{\alpha N}} DL_{\max}(N) \quad (25)$$

19

20 Where DL<sub>max</sub>(S), DL<sub>max</sub> (N), and DL<sub>min</sub>(N).are based on the critical load within a sensitive  
 21 areas that protects a specified percentile (e.g. 95%) of water bodies in the area.

22 Note that  $C_{\min}(NO_y)$  is a conditional function determined by the relationship between  
 23 total nitrogen buffering capacity in an ecosystem and the amount of reduced nitrogen deposition.  
 24 When reduced nitrogen deposition exceeds the buffering capacity of an ecosystem, then all  
 25 atmospheric oxidized nitrogen contributes to acidification. When reduced nitrogen deposition is  
 26 less than the buffering capacity of an ecosystem, then some amount of NO<sub>y</sub> is buffered (i.e. is  
 27 reflected in  $C_{\min}(NO_y)$ ) but that amount reflects the contribution of NH<sub>x</sub> to total nitrogen (the  
 28 amount of buffering capacity used up by reduced nitrogen). In this case, some fraction of the  
 29 atmospheric oxidized nitrogen may not contribute to acidification.

1 Recall that these three variables are conditional on the chosen level of AAPI, and reflect  
2 the depositional loadings that are associated with an equivalent level of ANC, e.g. for an AAPI  
3 of 50, the  $DL_{\max}(S)$ ,  $DL_{\max}(N)$ , and  $DL_{\min}(N)$  are associated with an ANC of 50. Also recall that  
4  $DL_{\max}(S)$  for a given ANC is a function of the “natural” flux of base cations to a watershed,  
5 runoff, and the amount of sulfur retention within a waterbody;  $DL_{\min}(N)$  is the minimum amount  
6 of deposition of total nitrogen ( $NHx + NOx$ ) that catchment processes can effectively remove  
7 without contributing to the acidic balance; and  $DL_{\max}(N)$  for a given ANC is a function of  
8  $DL_{\min}(N)$  and the “natural” flux of base cations to a watershed, runoff, and the amount of  
9 nitrogen retention within a waterbody, assuming S is zero. In our framework,  $DL_{\min}(N)$  is  
10 calculated from the FAB critical load modeling (equation 5 from Attachment A of the REA) or  
11 estimated through measured or modeled values of total nitrogen deposition and nitrate leaching.

12 As discussed in sections 5.3, the specific estimation of  $\left[ \frac{1}{Q} \cdot N_{eco} + [BC]_o^* \right]_{\%eco}$ ,  $T_{NOy}$ ,  
13  $T_{SOx}$  and  $NHx$  in a specific sensitive area will depend on the spatial scale of the sensitive area.  
14 Sensitivity can be assessed at the level of individual catchments, however, this presents practical  
15 limitations for establishing meaningful standards, as there are thousands of catchments within the  
16 U.S. Binning classes of sensitivity within larger spatial areas can provide a more manageable set  
17 of values of  $\left[ \frac{1}{Q} \cdot N_{eco} + [BC]_o^* \right]_{\%eco}$ ,  $T_{NOy}$ ,  $T_{SOx}$  and  $NHx$ . These parameters can be estimated in  
18 several ways for the larger spatial areas. Mean or median values can be generated across  
19 catchments, however, this would lead to parameter estimates that do not reflect conditions in the  
20 more sensitive lakes in the region. Alternatively, in order to provide a desired level of protection  
21 in these larger defined spatial areas, estimates based on higher percentiles of the distributions of  
22 parameters across catchments can be generated, e.g. the 75th or 95th percentile values of of  
23  $\left[ \frac{1}{Q} \cdot N_{eco} + [BC]_o^* \right]_{\%eco}$ ,  $T_{NOy}$ ,  $T_{SOx}$  and  $NHx$  could be used to provide protection for the more  
24 vulnerable aquatic ecosystems, however this would potentially lead to over-protection for less  
25 vulnerable ecosystems in the area. The Administrator may consider the balance between  
26 protection of particularly sensitive ecosystems and the overall protection for ecosystems in an  
27 area as an important element to consider in making decisions about the target level of ANC and  
28 the percent of aquatic ecosystems within an area targeted to achieve the specified ANC level.

1  
2 **5.4 Options for specifying the targets for the ecological indicator for aquatic**  
3 **acidification (ANC)**  
4

5 As discussed earlier in this chapter, ANC is the ecological indicator best suited to reflect  
6 the sensitivity of aquatic ecosystems to acidification (5.3). ANC is an indicator of the effects  
7 expected to occur given the natural buffering capacity of an ecosystem and the loadings of  
8 nitrogen and sulfur resulting from atmospheric deposition. **As noted by CASAC in their**  
9 **review of the first draft of this Policy Assessment, “information on levels of ANC**  
10 **protective to fish and other aquatic biota has been well developed and presents probably**  
11 **the lowest level of uncertainty in the entire methodology.”**

12 A target ANC limit is an important starting point in deciding the appropriate level of  
13 AAPI, and is a necessary component in determining the critical loads for N and S from which the  
14 tradeoff curves for ambient NO<sub>y</sub> and SO<sub>x</sub> are derived. In fact, the target ANC together with the  
15 selected percent of waterbodies to be protected to that target ANC (see section 5.4) uniquely  
16 determines the set of critical loads of N and S across the set of acid sensitivity categories.

17 **In reaching staff conclusions regarding the range of target ANC levels that is**  
18 **appropriate to evaluate further in the context of setting a NO<sub>x</sub> and SO<sub>x</sub> standard with the AAPI**  
19 **form, considering effects on public welfare due to aquatic acidification, we use three types of**  
20 **information: 1) direct information on levels of biological impairment related to**  
21 **alternative levels of ANC (and related levels of pH), 2) target ANC values and their**  
22 **rationales as identified by states or regions in setting critical loads to protect regional water**  
23 **bodies, and 3) information on ecosystem service losses associated with alternative ANC**  
24 **values.**

25 In addition, we evaluate risk-based information derived from the Risk Assessment,  
26 focused on case studies of acid sensitive lakes and streams in the Adirondacks and  
27 Shenendoahs. As part of our risk-based considerations, we have considered estimates of  
28 risks under current conditions in which current NO<sub>x</sub> and SO<sub>x</sub> air quality standards are met  
29 in the case study areas.

1 It is important to note that the choice of a target ANC level starts with an  
2 evaluation of the conditions that exist if a certain ANC occurs in fact. There is a  
3 significant body of science to inform that evaluation, as discussed below. However the  
4 development of a NAAQS based on the AAPI form does not assume or guarantee that  
5 any specific ANC level will in fact occur for any specific body of water. A critical issue for  
6 the Administrator to address in setting a NAAQS based on the AAPI is to consider and  
7 weigh the varying degrees of uncertainty in establishing the elements of the AAPI. These  
8 uncertainties impact the likelihood that a specific AAPI standard would in fact achieve a  
9 target ANC level for a specified percentage of a population of water bodies. Thus it is  
10 appropriate to first look at target ANC from the perspective of what we understand the  
11 impacts would be if a certain ANC level were realized. Hence, the discussion below  
12 focuses on evaluation of the ecological impacts at various target ANC levels, assuming the  
13 ANC level occurs in fact. Judgments made based on that evaluation then become the  
14 foundation for further judgments that would be made in the process of establishing the  
15 specific level of an AAPI, taking into account the degree of uncertainty in projecting the  
16 actual ANC levels that would result from any specific APPI standard.

17

#### 18 **5.4.1 What levels of impairment are related to alternative target levels of ANC?**

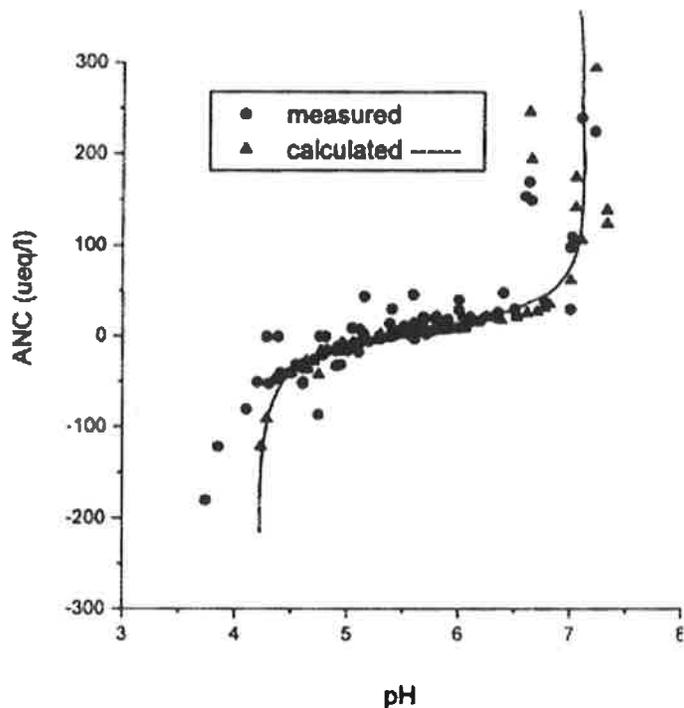
19 As discussed in Chapters 2, 3, and 4, specific levels of ANC are associated with differing  
20 levels of risk of ecosystem impairment, with higher levels of ANC resulting in lower risk of  
21 ecosystem impacts, and lower levels resulting in risk of both higher intensity of impacts and a  
22 broader set of impacts. While ANC is not the causal agent determining biological effects in  
23 aquatic ecosystem<sup>3</sup>, it is a useful metric for determining the level at which a water body is  
24 protected against risks of acidification. There is a direct correlation between ANC and pH  
25 levels, which, along with dissolved aluminum, are more closely linked to the biological causes of  
26 ecosystem response to acidification.

---

<sup>3</sup> Biological effects are primarily attributable to a combination of low pH and high inorganic Al concentration. Such conditions occur more frequently during rainfall and snowmelt that cause high flows of water and less commonly during low-flow conditions, except where chronic acidity conditions are severe. Higher ANC values are generally associated with lower risk of low pH during rainfall and snowmelt events.

1           Because there is a direct correlation between ANC and pH levels, we can inform  
2 the selection of target ANC in part through information on effects of pH as well as direct  
3 studies of ANC. Figure 5-23 demonstrates the relationship between the measure of risk to  
4 the aquatic ecosystems, ANC, and pH, the causal indicator of effect in aquatic ecosystems  
5 (Chapter 2). Levels of pH are closely associated with ANC in the pH range of 4.5 to 7.  
6 Within this range, higher ANC levels are associated with higher pH levels. At a pH level of  
7 4.5, further reductions in ANC appear to be uncorrelated with pH, as pH levels remain at  
8 4.5 while ANC values fall substantially. Likewise, at a pH value of 7, ANC values  
9 continue to increase with no corresponding increase in pH. As pH is the primary causal  
10 indicator of acidification related effects, this suggests that ANC values below  $-50 \mu\text{eq/L}$   
11 (the apparent point on the function where pH reaches a minimum) are not likely to result  
12 in further damage, while ANC values above  $50 \mu\text{eq/L}$  (the apparent point on the function  
13 where pH reaches a maximum) are not likely to confer additional protection. As a result,  
14 our focus will be on ANC values in the range of  $-50$  to  $50 \mu\text{eq/L}$ .

15



1  
 2 **Figure 5-23. Relationship between ANC and pH levels (reproduced from Bi et al, 2001)**

3  
 4        Within this ANC range, the role of ANC is to protect against the likelihood of  
 5 decreased pH (and associated increases in Al). Gerritson et al (1996) demonstrate that  
 6 the probability of pH decreasing to a given threshold level is affected by the ANC level. In  
 7 general, the higher the ANC, the lower the probability of decreasing to the threshold.  
 8 Figure 5-24, reproduced from Gerritson et al (1996) shows that for two common fish  
 9 species, the probability of exposure to pH levels below threshold values declines as ANC  
 10 levels increase. This suggests that greater protection against threshold values of pH that cause  
 11 biological damage would be achieved by target ANC values set high enough to have a low  
 12 probability of the system going below a threshold pH level.

13  
 14

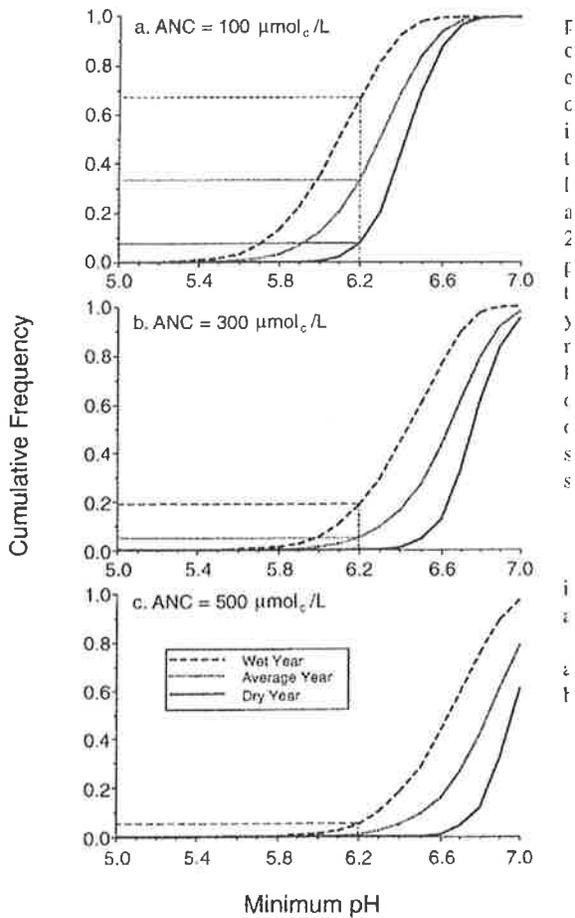


FIG. 5. Cumulative distributions of minimum pH event for streams with ANC = 100, 300, and 500  $\mu\text{mol}_c/\text{L}$ , respectively, during April in wet, average, and dry years.

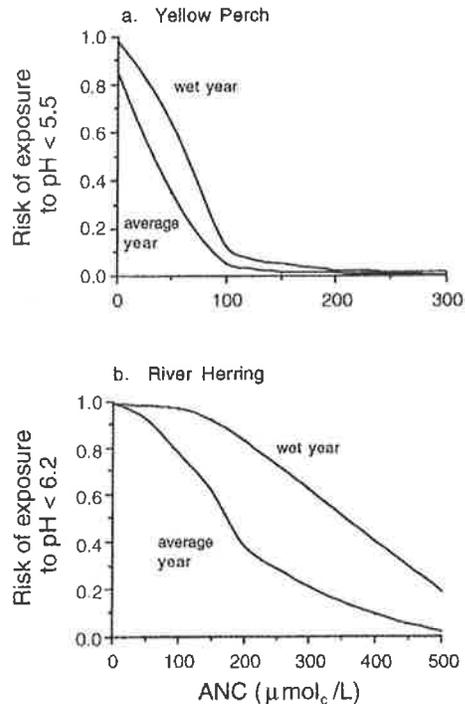


FIG. 6. Risk of exposure to one or more streamwater pH depressions below critical during average and dry years. (a) Yellow perch. (b) River herring (blueback herring and alewife).

1  
2 **Figure 5-24 Summary of results from Gerritson et al 1996. Blue back herring larval**  
3 **mortality occurs at pH 6.2. The higher the ANC the less likely it is that the pH will dip below**  
4 **6.2** Source Gerritsen et al. 1996

5  
6 Damages to aquatic ecosystems can be measured using several different metrics,  
7 including health of fish populations (measured using the condition factor, an index of fish length  
8 and weight which indicates loss in available energy due to acidification), biodiversity (e.g.  
9 number of fish species), and community structure (e.g. ratios of higher trophic level to lower  
10 trophic level species). The next two sections discuss the evidence for a range of damage types  
11 occurring at different levels of pH and ANC.

1 *Evidence of effects due to low pH levels:*

2           Significant harm to sensitive aquatic species has been observed at pH levels below 6.  
3 Normal stream pH levels with little to no toxicity ranges from 6 to 7 (MacAvoy et al, 1995).  
4 Baker et al (1990) observed that “lakes with pH less than approximately 6.0 contain significantly  
5 fewer species than lakes with pH levels above 6.0”. As noted in Chapter 2, typically at pH <4.5  
6 and an ANC <0 µeq/L, complete to near-complete loss of many taxa of organisms occur,  
7 including fish and aquatic insect populations, whereas other taxa are reduced to only acidophilic  
8 species.

9           Additional evidence can help refine the understanding of effects occurring at pH levels  
10 between 4.5 and 6. When pH levels are below 5.6, relatively lower trout survival rates were  
11 observed in the Shenendoah National Park. In field observations, when pH levels dropped to 5,  
12 mortality rates went to 100 percent. (Bulger et al, 2000). At pH levels ranging from 5.4 to 5.8,  
13 cumulative mortality continues to increase. Several studies have shown that trout exposed to  
14 water with varying pH levels and fish larvae showed increasing mortality as pH levels decrease.  
15 In one study almost 100 percent mortality was observed at a pH of 4.5 compared to almost 100  
16 percent survival at a pH of 6.5. Intermediate pH values (6.0, 5.5) in all cases showed reduced  
17 survival compared with the control (6.5), but not by statistically significant amounts (ISA  
18 3.2.3.3). One study (Woodward, 1991) concluded that the threshold for effects of acidity on  
19 greenback cutthroat trout in the absence of inorganic Al was pH 5.0.

20           One important indicator of acid stress is increased fish mortality. Table 5-9 summarizes  
21 the evidence of mortality related to different levels of pH for a number of fish species.

1  
2

**Table 5-9. Summary of Fish Mortality Response to pH**

Source: EPA, 2008 (ISA)

<b>Mortality Endpoint</b>	<b>Authors</b>	<b>Species</b>	<b>pH Level</b>	<b>Notes</b>
<b>Increased Mortality</b>	Johnson et al. (1987)	Blacknose dace, creek chub	5.9 - 6.0	In situ bioassay with early life stages in Adirondack surface waters
		Brook trout	4.8 - 5.1	
	Holtze and Hutchinson (1989)	Common shiner	5.4 - 6.0	Laboratory exposure of early life stages to pH and Al.
		Lake whitefish, white sucker, walleye	5.1 - 5.2	
		Smallmouth bass	4.8	
	Johansson et al. (1977)	Atlantic salmon	5.0	Laboratory tests with eggs exposed to low pH, no Al.
		Brown trout	4.5 - 5.0	
		Brook Trout	4.5	
	Swenson et al. (1989)	Black crappie	5.5	Laboratory tests with early life stages exposed to pH and Al.
		Rock bass	5.0	
		Yellow perch, largemouth bass	4.5	
	Mills et al. (1987)	Fathead minnow	5.9	Whole-lake treatment (fish population recruitment failure)
		Slimy sculpin	5.6 - 5.9	
		Lake Trout	5.6	
		Pearl dace	5.1	
White sucker		5.0 - 5.1		
<b>&gt;50% larval mortality</b>	Buckler et al. (1987)	Striped bass	6.5	Lab bioassay
	Klauda et al. (1987)	Blueback herring	5.7	Lab bioassay
	Kane and Rabeni (1987)	Smallmouth bass	5.1	Lab bioassay
<b>embryo survival</b>				
Significant decrease	McCormick et al. (1989)	Fathead minnow	6.0	Lab bioassay
>50% embryo mortality	Holtze and Hutchinson (1989)	Common shiner	5.4	Lab bioassay
Substantial reduction	Baker and Schofield (1980)	White sucker	5.2	Lab bioassay

3

1  
 2 The response of fish to pH is not uniform across species. A number of synoptic surveys  
 3 indicated loss of species diversity and absence of several fish species in the pH range of 5.0 to  
 4 5.5. If pH is lower, there is a greater likelihood that more fish species could be lost without  
 5 replacement, resulting in decreased richness and diversity. In general, populations of salmonids  
 6 are not found at pH levels less than 5.0, and smallmouth bass (*Micropterus dolomieu*)  
 7 populations are usually not found at pH values less than about 5.2 to 5.5. From Table 5-10, only  
 8 one study showed significant mortality effects above a pH of 6, while a number of studies  
 9 showed significant mortality when pH levels are at or below 5.5.

10  
 11 **Table 5-10 Threshold response of increased mortality of fish to low pH listed from least**  
 12 **sensitive to most sensitive**

Study	Species	Increased Mortality Threshold, pH	Study Conditions
Johnson et al. (1987)	Blacknose dace, creek chub	5.9 - 6.0	In situ bioassay with early life stages in Adirondack surface waters
	Brook trout	4.8 - 5.1	
Holtze and Hutchinson (1989)	Common shiner	5.4 - 6.0	Laboratory exposure of early life stages to pH and Al.
	Lake whitefish, white sucker, walleye	5.1 - 5.2	
	Smallmouth bass	4.8	
Johansson et al. (1977)	Atlantic salmon	5.0	Laboratory tests with eggs exposed to low pH, no Al.
	Brown trout	4.5 - 5.0	
	Brook Trout	4.5	
Swenson et al. (1989)	Black crappie	5.5	Laboratory tests with early life stages exposed to pH and Al
	Rock bass	5.0	
	Yellow perch, largemouth bass	4.5	
Mills et al. (1987)	Fathead minnow	5.9	Whole-lake treatment (fish population recruitment failure)
	Slimy sculpin	5.6 - 5.9	
	Lake Trout	5.6	
	Pearl dace	5.1	
	White sucker	5.0 - 5.1	

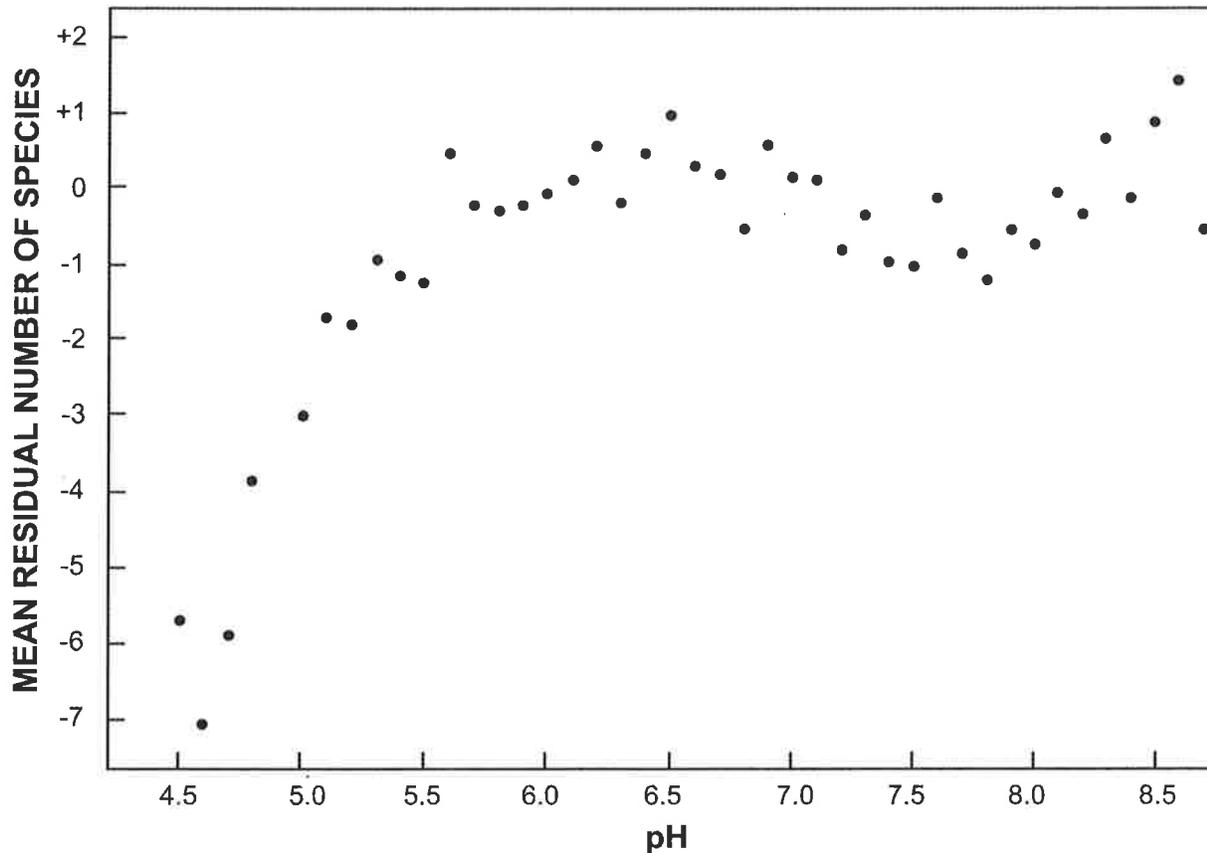
13  
 14 Source: Baker et al 1990, reproduced from the ISA, Table B-23.

1 The highest pH threshold for any of the studies reported in Table 5-10 is 6.0, suggesting that pH  
2 above 6.0 is protective against mortality effects for most species. Most thresholds are in the  
3 range of pH of 5.0 to 6.0, which suggests that a target pH should be no lower than 5.0.  
4 Protection against mortality in some recreationally important species such as lake trout (pH  
5 threshold of 5.6) and crappie (pH threshold of 5.5), combined with the evidence of effects on  
6 larval and embryo survival suggests that pH levels greater than 5.5 should be targeted to provide  
7 protection against mortality effects throughout the lifestages of fish.

8 Non-lethal effects have been observed at pH levels as high as 6. A study in the  
9 Shenendoah National Park found that the condition factor, a measure of fish health expressed as  
10 fish weight/length<sup>3</sup> multiplied by a scaling constant, is positively correlated with stream pH  
11 levels, and that the condition factor is reduced in streams with a pH of 6.0 (ISA 3.2.3.3).

12 Biodiversity is another indicator of aquatic ecosystem health. As discussed in Chapter 2,  
13 a key study in the Adirondacks found that lakes with a pH of 6.0 had only half the potential  
14 species of fish (27 of 53 potential species). There is often a positive relationship between pH and  
15 number of fish species, at least for pH values between about 5.0 and 6.5, or ANC values between  
16 about 0 to 100 µeq/L (Bulger et al., 1999; Cosby et al., 2006; Sullivan et al., 2006). Such  
17 observed relationships are complicated, however, by the tendency for smaller lakes and streams,  
18 having smaller watersheds, to also support fewer fish species, irrespective of acid-base  
19 chemistry. This pattern may be due to a decrease in the number of available niches as stream or  
20 lake size decreases. Nevertheless, fish species richness is relatively easily determined and is one  
21 of the most useful indicators of biological effects of surface water acidification.

22 In a study of Ontario lakes, Matuszek and Beggs (1988) found that the number of fish  
23 species is positively correlated with pH, with a clear loss of species starting at pH levels less than  
24 or equal to 5.5. This relationship is displayed in Figure 5-25.



1  
2

**Figure 5-25. Mean residual number of species per lake for lakes in Ontario, by pH interval. The residual number of species for a lake is the deviation of the observed number from the number predicted by lake area.**

Source: Matuszek and Beggs (1988).

3

4 A study in the Adirondacks found that among the studied lakes with fish, there was an  
5 unambiguous relationship between the number of fish species and lake pH, ranging from about  
6 one species per lake for lakes having pH less than 4.5 to about six species per lake for lakes  
7 having pH higher than 6.5 (Baker et al., 1990b; Driscoll et al., 2001).

8 Biological responses at differing pH levels are summarized in Table B-13 of the ISA,  
9 reproduced here in Table 5-11.

10

11

12

13

1 **Table 5-11 General summary of biological changes anticipated with surface water**  
 2 **acidification, expressed as a decrease in surface water pH.**

pH Decrease	General Biological Effects
6.5 to 6.0	<p>Small decrease in species richness of plankton and benthic invertebrate communities resulting from the loss of a few highly acid-sensitive species, but no measurable change in total community abundance or production.</p> <p>Some adverse effects (decreased reproductive success) may occur for highly acid-sensitive fish species (e.g., fathead minnow, striped bass).</p>
6.0 to 5.5	<p>Loss of sensitive species of minnows and dace, such as fathead minnow and blacknose dace; in some waters, decreased reproductive success of lake trout and walleye, which are important sport fish species in some areas.</p> <p>Visual accumulation of filamentous green algae in the near-shore zone of many lakes and in some streams.</p> <p>Distinct decrease in species richness and change in species composition of plankton and benthic invertebrate communities, although little if any change in total community abundance or production.</p> <p>Loss of some common invertebrate species from zooplankton and benthic communities, including many species of snails, clams, mayflies, and amphipods, and some crayfish.</p>
5.5 to 5.0	<p>Loss of several important sport fish species, including lake trout, walleye, rainbow trout, and smallmouth bass, as well as additional nongame species such as creek chub.</p> <p>Further increase in the extent and abundance of filamentous green algae in lake near-shore areas and streams.</p> <p>Continued shift in species composition and decline in species richness of plankton, periphyton, and benthic invertebrate communities; decreases in total abundance and biomass of benthic invertebrates and zooplankton may occur in some waters.</p> <p>Loss of several additional invertebrate species common in surface waters, including all snails, most species of clams, and many species of mayflies, stoneflies, and other benthic invertebrates.</p> <p>Inhibition of nitrification.</p>

pH Decrease	General Biological Effects
5.0 to 4.5	<p>Loss of most fish species, including most important sport fish species such as brook trout and Atlantic salmon. A few fish species are able to survive and reproduce in water below pH 4.5 (e.g., central mudminnow, yellow perch, and in some waters, largemouth bass).</p> <p>Measurable decline in the whole-system rates of decomposition of some forms of organic matter, potentially resulting in decreased rates of nutrient cycling.</p>

1  
2 Evidence of changes in biological endpoints in response to changes in pH indicates that  
3 while the time to full response varies, and the pH level at which recovery occurs varies, there is  
4 clear evidence that increasing pH levels leads to improvements in a number of measures of  
5 ecosystem health. In a study in Ontario, there was varied response to increases in pH across  
6 studied lakes, with little increase in phytoplankton diversity in one lake as pH changed from 5.0  
7 to 5.8 but a strong recovery of diversity at pH above 6 (Findlay and Kasian, 1996), while in  
8 another lake, profound change began at pH 5.5.

9 Fish populations have recovered in acidified lakes when the pH and ANC have been  
10 increased through liming or reduction of acidifying deposition (Beggs and Gunn, 1986; Dillon et  
11 al., 1986; Gunn et al., 1988; Hultberg and Andersson, 1982; Keller and Pitblado, 1986; Kelso  
12 and Jeffries, 1988; Raddum et al., 1986).

13 In considering the range of pH values to evaluate further in the context of impacts on  
14 public welfare, the overall weight of evidence regarding the full range of biological effect  
15 indicators, including mortality, fish health, and biodiversity supports further consideration of a  
16 target pH level of no less than 5.5, with some support for pH target levels of 6 to 6.5.

17  
18 *Evidence of effects directly or indirectly related to low ANC levels:*

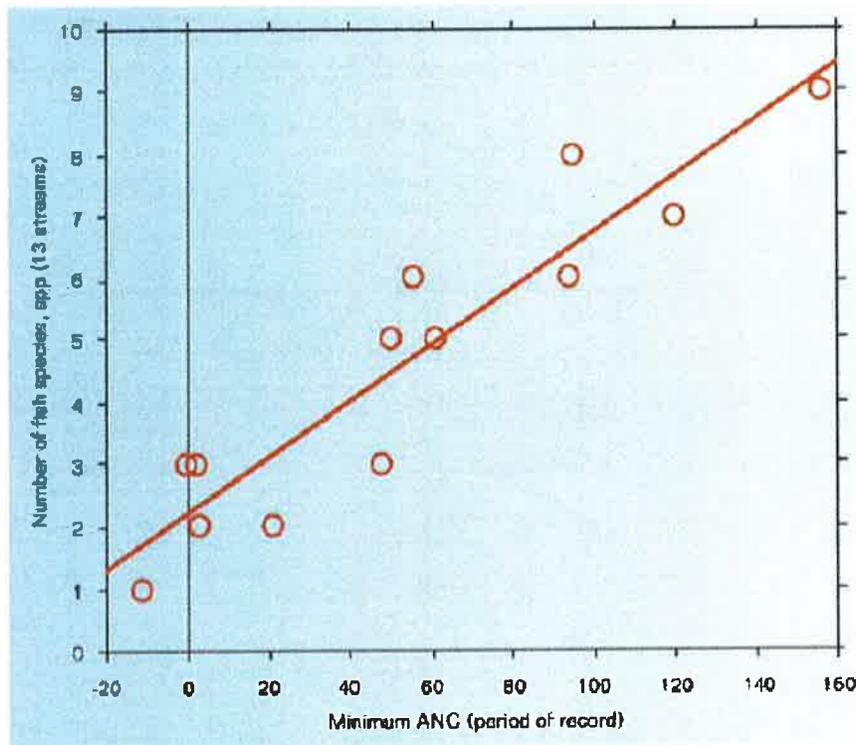
19 The number of fish species present in a waterbody has been shown to be positively  
20 correlated with the ANC level in the water, with higher values supporting a greater richness and  
21 diversity of fish species (Figure 5-26 and 5-27). The diversity and distribution of phyto-  
22 zooplankton communities are also positively correlated with ANC.

23 When comparing lower ANC to higher ANC streams, the lower ANC streams  
24 demonstrated severe mortality in young fish due to a sharp drop in pH and increase in Al

1 concentrations. The higher ANC stream maintained a pH level greater than 6.6 with  
2 correspondingly low Al concentrations (MacAvoy et al 1995). In streams with chronically low  
3 ANC, pH levels were in the range of 5.3 to 5.6, and a steady decline in trout populations was  
4 observed.

5 Within the range where ANC is the limiting indicator, (e.g. ANC range from -50 to 100  
6  $\mu\text{eq/L}$ ) the relationship between ANC and ecosystem impacts ranges from linear to non-linear,  
7 with a sigmoidal shape. Evidence shows a linear relationship in streams in the Eastern U.S., and  
8 a non-linear relationship in lakes in the Northeastern U.S. (Figures 5-26 and 5-27).

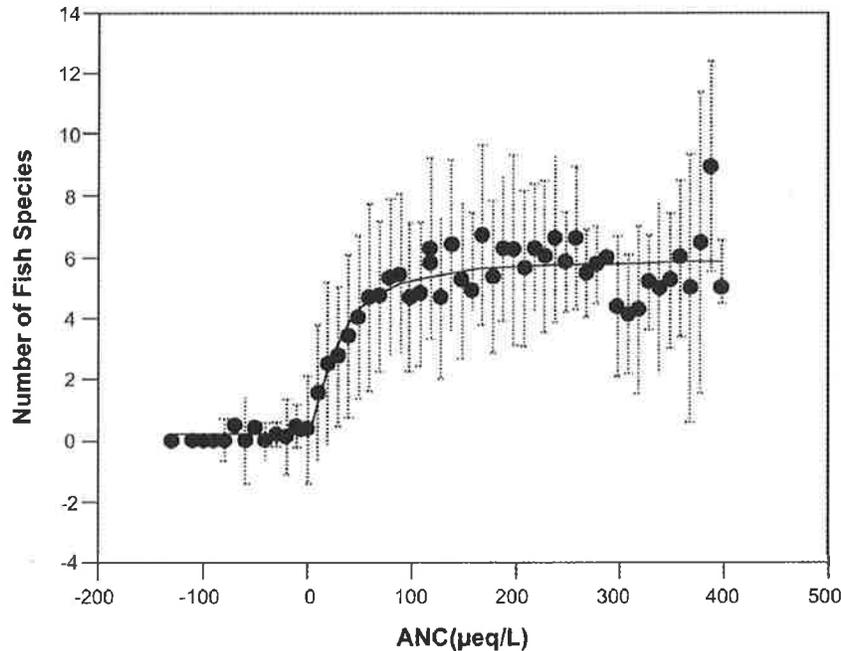
9



10

11 **Figure 5-26 Relationship between ANC and number of fish species present in aquatic**  
12 **freshwater ecosystems in Shenandoah National Park** (Source: Arthur Bulger,  
13 University of Virginia, reproduced from NAPAP, 2005.)

14



Source: Sullivan et al. (2006)

**Figure 5-27. Number of fish species per lake versus acidity status, expressed as ANC, for Adirondack lakes.** The data are presented as mean (filled circles) and range (bars) of species richness within 10 µeq/L ANC categories, based on data collected by the Adirondack Lakes Survey Corporation.

1 On average, the fish species richness is lower by one fish species for every 21 µeq/L  
 2 decrease in ANC in Shenandoah National Park streams (ISA 3.2.3.4).

3 For freshwater systems, ANC levels can be grouped into five major classes: <0, 0–20,  
 4 20–50, 50–100, and >100 µeq/L, with each range representing a probability of ecological  
 5 damage to the community. The five categories of ANC and expected ecological effects are  
 6 described Table 2-1 in Chapter 2 and are supported by a large body of research completed  
 7 throughout the eastern United States (Sullivan et al., 2006).

8 In a study in Maryland, Gerritsen et al. (1996) found that at an ANC less than 40 to 70  
 9 µeq/L, Yellow perch had a risk of exposure to its critical pH =5.5 of greater than 50 percent.  
 10 Figures from that study, reproduced in Figure 5-24 above, show the relationships between ANC  
 11 and the probability of exceeding benchmark pH levels that have been associated with mortality  
 12 in the Yellow perch and River herring. These figures demonstrate two important principles, 1)  
 13 as ANC levels fall, the distribution of pH shifts to the left, with increasing probability of low pH

1 levels, and 2) as ANC levels fall, the probability of exceeding mortality thresholds increases, but  
2 this relationship varies by fish species.

3 The specific relationship between ANC and the probability of exceeding benchmark pH  
4 levels varies by water body and fish species. However, based on Figure 5-23, ANC levels  
5 matching the target pH levels of 5.5 to 6.5 discussed in the previous section range from  
6 approximately 0 to 50  $\mu\text{eq/L}$ . In considering the range of ANC values to evaluate further in the  
7 context of impacts on public welfare, the overall evidence on effects at lower ANC levels  
8 described in Table 2-1, supports further consideration of a range of ANC from 20 to 50  $\mu\text{eq/L}$ .  
9 While Figure 5-23 suggests some probability that ANC levels down to 0  $\mu\text{eq/L}$  are correlated  
10 with pH levels of 5.5, the specific relationship between ANC and fish species diversity shown in  
11 Figure 5-27 indicates that at an ANC of 0  $\mu\text{eq/L}$ , there is significant damage to ecosystems with  
12 almost complete loss of fish species, which indicates that an ANC of 0  $\mu\text{eq/L}$  is not an  
13 appropriate target level to protect against significant damage to ecosystems.

#### 14 15 *Evidence and Risk Based Conclusions regarding target ANC levels*

16 ANC values less than or equal to 0  $\mu\text{eq/L}$  are chronically acidic and can lead to complete  
17 loss of species, and major changes in the ability of waterbodies to support diverse biota,  
18 especially in water bodies that are highly sensitive to episodic acidification. Biota are generally  
19 not harmed when ANC values are  $>100 \mu\text{eq/L}$ , due to the low probability that pH levels will be  
20 below 7. In the Adirondacks, the number of fish species also peaks at ANC values  $>100 \mu\text{eq/L}$ .  
21 This suggests that at ANC greater than 100, little risk from acidification exists in many aquatic  
22 ecosystems. At ANC levels below 100  $\mu\text{eq/L}$ , overall health of an aquatic community can be  
23 maintained; however, fish fitness and community diversity begin to decline. At ANC levels  
24 between 100 and 50  $\mu\text{eq/L}$ , the likelihood that fitness of sensitive species (e.g., brook trout,  
25 zooplankton) will begins to decline is increased. When ANC concentrations are  $<50 \mu\text{eq/L}$ , the  
26 probability of acidification increases substantially, and negative effects on aquatic biota are  
27 observed, including large reductions in diversity of fish species, and changes in health of fish  
28 populations, affecting reproductive ability and fitness. ANC levels below 20  $\mu\text{eq/L}$  are generally  
29 associated with high probability of low pH, leading to death or loss of fitness of biota that are  
30 sensitive to acidification. (**ISA 5.2.2.1 and REA 5.2.1.2**). At these levels, during episodes of  
31 high acidifying deposition, brook trout populations may experience lethal effects. In addition, the

1 diversity and distribution of zooplankton communities decline sharply at ANC levels below 20  
2  $\mu\text{eq/L}$ . Overall, there is little uncertainty that significant effects on aquatic biota are occurring at  
3 ANC levels below 20  $\mu\text{eq/L}$ .

4       Based on the field data from the Adirondacks and Shenendoah case study areas, ANC  
5 levels less than 50  $\mu\text{eq/L}$  are adverse to ecosystem health, and are likely to lead to reductions in  
6 ecosystem services related to recreational fishing. However, the types of effects, specific  
7 species, and prevalence of effects across water bodies in the U.S. is more uncertain at ANC  
8 levels between 20 and 50  $\mu\text{eq/L}$ . ANC levels between 50 and 100  $\mu\text{eq/L}$  are potentially adverse  
9 to ecosystem health, and may result in losses in ecosystem services, but the effects are less  
10 severe and greater uncertainty exists as to the magnitude of ecosystem service impacts.

11       **Consideration of the appropriate levels of ANC to target in the standard to reduce**  
12 **the likelihood of effects from aquatic acidification can be based upon the above presented**  
13 **categories of aquatic status in Table 2-1. Using this information as well as information**  
14 **provided by both the ISA and REA, the lowest two categories (0 and <20  $\mu\text{eq/L}$ ) would**  
15 **appear inadequate to protect against catastrophic loss of ecosystem function. While**  
16 **ecological effects occur at ANC levels below 50  $\mu\text{eq/L}$ , the degree and nature of those**  
17 **effects is less significant than at levels below 20  $\mu\text{eq/L}$ . Levels between 50 and 100  $\mu\text{eq/L}$**   
18 **would provide additional protection; however, uncertainties regarding the additional**  
19 **reduction in adverse welfare effects are much larger for target ANC levels above 50  $\mu\text{eq/L}$ .**

20       The target ANC level specified in designing the standard is only one part in  
21 determining the overall protectiveness of the standard. The degree of protectiveness is  
22 based on all elements of the standard, including the target ANC, the size of the spatial  
23 areas over which the standard is applied, the percent of aquatic ecosystems targeted within  
24 a spatial area that is selected by the Administrator in light of the selected ANC level, the  
25 atmospheric indicator, the calculated values for the deposition transformation ratios ( $T_{\text{NO}_x}$   
26 and  $T_{\text{SO}_x}$ ), and the calculated value for reduced nitrogen deposition ( $\text{NH}_x$ ). There are  
27 widely varying degrees of uncertainty associated with all of these elements, some being

1 much more certain and others being much less certain. The specified target ANC  
2 level is a crucial part in determining the adequacy of protection provided by an AAPI, but  
3 it is the overall design and content of the standard that must be considered in judging the  
4 adequacy of protection it provides.

5 Consideration of the target ANC should also reflect that an adequate level of ANC  
6 should protect against episodic as well as long term effects. Selecting a higher chronic  
7 ANC level can provide greater protection against short term peaks in acidification. In  
8 addition, selection of ANC values in the lower end of the range of 20 to 50  $\mu\text{eq/L}$   
9 provides less protection against these short term episodic effects. Selection of target ANC  
10 values in the upper end of the range from 20 to 50  $\mu\text{eq/L}$  provides additional protection  
11 against episodic peaks in acidification.

12 When considering a standard to protect against aquatic acidification, it is necessary  
13 to take into account both the time period desired for recovery as well as the potential of  
14 recovery. Ecosystems become adversely impacted by acidifying deposition over long  
15 periods of time and have variable time frames and abilities to recover from such  
16 perturbations. Modeling presented in the REA (REA Section 4.2.4) shows the estimated ANC  
17 values for Adirondack lakes and Shenandoah streams under pre-acidification conditions and  
18 indicates that for a small percentage of lakes and streams, natural ANC levels would have  
19 been below 50  $\mu\text{eq/L}$ . Therefore, for these waterbodies, no reduction in input is likely to  
20 achieve an ANC of 50  $\mu\text{eq/L}$  or greater. Conversely, for some lakes and streams the level  
21 of perturbation from long periods of acidifying deposition has resulted in very low ANC  
22 values compared to estimated natural conditions. For such waterbodies, the time to  
23 recovery would be largely dependent on future inputs of acidifying deposition.

24 The concept of target ANC is based on the long-term response of aquatic  
25 ecosystems. The time required for a waterbody to achieve the target ANC given a  
26 decrease in emissions such that the critical load for that target ANC is not exceeded is  
27 often decades if not centuries. In recognition of the potential desire to achieve the target  
28 ANC in a shorter time frame, the concept of target loads had been developed. Target  
29 loads represent the depositional loading that is expected to achieve a particular level of the

1 ecological indicator by a given time. Similarly, one might also consider specifying a target  
2 ANC that will yield an intermediate ANC level within a specified time horizon. For  
3 example, if there is a desire to obtain an ANC of 20  $\mu\text{eq/L}$  by 2030, it might be necessary  
4 to use the depositional loadings equivalent to obtaining a target ANC of 50  $\mu\text{eq/L}$  which  
5 will ultimately be realized many years later. The depositional loading associated with the  
6 interim ANC target is called the target load. The implication of this is that an important  
7 component of the decision on the level of the AAPI is the timeframe in which the desired  
8 ANC value is to be achieved. There is a great deal of heterogeneity in the response time  
9 among waterbodies, and as such, specific quantification of the relationship between the  
10 critical loads and target loads is not possible.

11

#### 12 **5.4.2 Additional information on Target ANC levels**

13 A number of regional organizations, states, and international organizations have  
14 developed critical loads frameworks to protect against acidification of sensitive aquatic  
15 ecosystems. In considering the appropriate range of target ANC levels, it is informative to  
16 evaluate the target ANC levels selected by these different organizations, as well as the  
17 rationale provided in support of the selected levels. Chapter 3 provides a detailed  
18 discussion of how critical loads have been developed and used in other contexts. This  
19 section summarizes the specific target ANC values and their rationales.

20 The UNECE has developed critical loads in support of international emissions  
21 reduction agreements. As noted in Chapter 3, critical loads were established to protect  
22 95 percent of surface waters in Europe from an ANC less than 20  $\mu\text{eq/L}$ , based on  
23 protection of brown trout. Individual countries have set alternative ANC targets, for  
24 example, Norway targets an ANC of 30  $\mu\text{eq/L}$ , based on protection of Atlantic salmon.

25 Several states have established target ANC or pH values related to protection of  
26 lakes and streams from acidification. While recognizing that some lakes in the Adirondacks  
27 will have a naturally low pH, the state of New York has established a target pH value of  
28 6.5 for lakes that are not naturally below 6.5. As noted above this level is associated with  
29 an ANC value that is likely to be between 20 and 50  $\mu\text{eq/L}$  (or higher). Vermont has set

1 an explicit ANC target of 50  $\mu\text{eq/L}$ . Tennessee has established site specific target ANC  
2 values based on assessments of natural acidity, with a default value of 50  $\mu\text{eq/L}$  when  
3 specific data is not available.

4 Taken together, these policy responses to concerns about ecological effects  
5 associated with acidification indicate that target ANC values between 20 and 50  $\mu\text{eq/L}$   
6 have been selected by states and other nations to provide adequate protection of lakes and  
7 streams in some of the more sensitive aquatic ecosystems.

### 9 **5.4.3 Adversity of ecological impacts associated with alternative target ANC levels**

10 The point at which effects on public welfare become adverse is not specifically defined in  
11 the Clean Air Act. In Chapter 3 we explained that characterizing a known or anticipated adverse  
12 effect to public welfare is an important component of developing any secondary NAAQS.

13 According to the Clean Air Act, welfare effects include:

14 effects on soils, water, crops, vegetation, manmade materials, animals, wildlife,  
15 weather, visibility, and climate, damage to and deterioration of property, and  
16 hazards to transportation, as well as effect on economic values and on personal  
17 comfort and well-being, whether caused by transformation, conversion, or  
18 combination with other air pollutants (CAA, Section 302(h)).  
19

20 While the text above lists a number of welfare effects, these effects do not define public  
21 welfare in and of themselves. Consideration of adversity in the context of the secondary  
22 NO<sub>x</sub> and SO<sub>x</sub> standards can be informed by information about losses in ecosystem  
23 services associated with acid deposition, and the potential economic value of those losses  
24 (Chapter 3).

25 Ecosystem service losses at alternative levels of ANC are difficult to enumerate.  
26 However, in general there are categories of ecosystem services (discussed in detail in Chapter 3)  
27 that are related to the specific ecosystem damages expected to occur at alternative ANC levels.  
28 Losses in fish populations due to very low ANC (below 20  $\mu\text{eq/L}$ ) are likely associated with  
29 significant losses in value for recreational and subsistence fishers. Many acid sensitive lakes are  
30 located in areas with high levels of recreational fishing activity. For example, in the  
31 Northeastern U.S., where nearly 8 percent of lakes are considered acidic, more than 9 percent of  
32 adults participate in freshwater fishing, with a value of \$5 billion in 2006. This suggests that

1 improvements in lake fish populations are likely associated with significant recreational fishing  
2 value.

3 Inland surface waters also provide cultural services such as aesthetic and existence value  
4 and educational services. To the extent that piscivorous birds and other wildlife are harmed by  
5 the absence of fish in these waters, hunting and birdwatching activities are likely to be adversely  
6 affected.

7 A case study of the value to New York residents of improving the health of lakes in the  
8 Adirondacks found significant willingness to pay for those improvements. When scaled to  
9 evaluate the improvement in lake health from achieving an ANC of 20 to 50  $\mu\text{eq/L}$ , the study  
10 implies benefits to the New York population of \$400 to \$900 million per year (in constant  
11 2007\$) for achievement of an ANC of 20  $\mu\text{eq/L}$ , and \$300 to \$800 million per year for achieving  
12 an ANC of 50  $\mu\text{eq/L}$ . The survey administered in this study recognized that participants were  
13 thinking about the full range of services provided by the lakes in question – not just the  
14 recreational fishing services. Therefore the estimates of WTP include resident's benefits for  
15 potential birdwatching and other ancillary services. These results are just for New York  
16 populations. If similar benefits exist for improvements in other acid sensitive lakes, the  
17 economic value to U.S. populations could be very substantial, suggesting that, at least by one  
18 measure of public welfare, impacts associated with ANC greater than 50  $\mu\text{eq/L}$  are adverse to  
19 public welfare.

20

## 5.5 Options for specifying the targets for the deposition metric

This section outlines the steps necessary to aggregate critical loads to form a deposition metric ( $DL_{\%ECO}$ ) for a broad geographic area at a desired level of protection. In this chapter, evaluate  $DL_{\%ECO}$  for several of the options for categorization of landscape sensitivity based on acid-sensitivity that are presented in section 5.3. The options include one  $DL_{\%ECO}$  for the entire population of lakes and streams at a given level of protection, creating two categories (sensitive and less-sensitive) with a distinct  $DL_{\%ECO}$  for each, and a cluster approach which creates multiple categories of sensitivity for which  $DL_{\%ECO}$  are calculated. Regardless of the method of aggregation, the question becomes how to choose a  $DL_{\%ECO}$  value to represent the population of CL for waterbodies. For the purpose of comparison, we have chosen to calculate the  $DL_{\%ECO}$  from the 90 percentile CL, 75 percentile CL and 50 percentile CL for each method of aggregation. Using this method, the target  $DL_{\%ECO}$  becomes that value which represents protection for 90%, 75% or 50% of the population of waterbodies for a desired level of ANC and would not offer the same degree of protection to those waterbodies with CL less than the chosen  $DL_{\%ECO}$ . While we would not expect the same degree of protection for these waterbodies, they would likely receive some benefit from the reduced deposition necessary to meet the selected  $DL_{\%ECO}$ . This chapter will therefore also present comparisons between the percentage of waterbodies that would be protected at given  $DL_{\%ECO}$  from an ANC <50  $\mu\text{eq/L}$  and those that would likely be protected from an ANC <20  $\mu\text{eq/L}$  at the same  $DL_{\%ECO}$ . These comparisons are described below.

Table 5.12 shows a comparison between the percent of waterbodies that would be protected from ANC<50 using 90%, 75% and 50%  $DL_{\%ECO}$  for the one population approach and those that, while not protected from an ANC <50  $\mu\text{eq/L}$  would be protected from an ANC<20  $\mu\text{eq/L}$ . The selection of the  $DL_{\%ECO}$  for ANC at 50  $\mu\text{eq/L}$  representing 90% of the waterbodies would likely protect 97% of all waterbodies from having an ANC<20  $\mu\text{eq/L}$ . If the 75%  $DL_{\%ECO}$  for ANC 50  $\mu\text{eq/L}$  was chosen, 83% of waterbodies would likely be protected from an ANC<20  $\mu\text{eq/L}$  and if the 50%  $DL_{\%ECO}$  was chosen only 56% of waterbodies would likely be protected

1 against an ANC <20  $\mu\text{eq/L}$ . This is an important distinction as severe degradation is likely to  
 2 occur in lakes and streams with ANC<20  $\mu\text{eq/L}$ .

3

**Table 5-12. Comparison of percentage protection from ANC values less than 50 and less than 20 using DL that result when the US is considered one population.**

Percentile protection	$DL_{\%ECO}$ ( $\mu\text{eq/m}^2/\text{yr}$ )	Total number of Sites in Analysis	Total Number of Sites protected from ANC <50	Total % Sites protected from ANC <50	Total Number of Sites protected from ANC <20	Total % Sites protected from ANC <20
90%	27	5280	4778	90	5145	97
75%	55	5280	3973	75	4394	83
50%	118	5280	2654	50	2947	56

4

5 Table 5.13 shows a comparison between the percent of waterbodies that would be  
 6 protected from ANC<50  $\mu\text{eq/L}$  using 90%, 75% and 50%  $DL_{\%ECO}$  s for subdividing the U.S. into  
 7 two acid-sensitivity categories and those that, while not protected from an ANC <50 would  
 8 likely be protected from an ANC<20  $\mu\text{eq/L}$ . **Sensitive Category:** The selection of the  $DL_{\%ECO}$   
 9 for ANC50  $\mu\text{eq/L}$  representing 90% of the sensitive waterbodies would likely protect 98% of all  
 10 waterbodies from having an ANC<20  $\mu\text{eq/L}$ . If the 75%  $DL_{\%ECO}$  for ANC 50  $\mu\text{eq/L}$  for sensitive  
 11 waterbodies was chosen, 84% of waterbodies would likely be protected from an ANC<20  $\mu\text{eq/L}$   
 12 and if the 50%  $DL_{\%ECO}$  were chosen only 57% of waterbodies would likely be protected against  
 13 an ANC <20  $\mu\text{eq/L}$ . **Less-sensitive Category:** The selection of the  $DL_{\%ECO}$  for ANC50  
 14 representing 90% of the non-sensitive waterbodies would likely protect 92% of all waterbodies  
 15 from having an ANC<20  $\mu\text{eq/L}$ . If the 75%  $DL_{\%ECO}$  for ANC 50  $\mu\text{eq/L}$  for sensitive waterbodies  
 16 was chosen, 77% of waterbodies would likely be protected from an ANC<20  $\mu\text{eq/L}$  and if the  
 17 50%  $DL_{\%ECO}$  were chosen only 52% of waterbodies would likely be protected against an ANC  
 18 <20  $\mu\text{eq/L}$ .

19

20

**Table 5-13. Comparison of percentage protection from ANC values less than 50 and less than 20 using DL that result when the US is divided into two categories, sensitive and less sensitive based on ANC data.**

	Percentile protection	$DL_{\%ECO}$ (meq/m <sup>2</sup> /yr)	Total number of Sites in Analysis	Total Number of Sites protected from ANC <50	Total % Sites protected from ANC <50	Total Number of Sites protected from ANC <20	Total % Sites protected from ANC <20
Sensitive	90%	26	4553	4104	90	4451	98
	75%	51	4553	3428	75	3841	84
	50%	106	4553	2284	50	2575	57
Less sensitive	90%	53	727	655	90	672	92
	75%	117	727	546	75	560	77
	50%	277	727	364	50	377	52

1

2           The sensitive category can be further subdivided into ecoregions (Option 2c in section  
3 5.3) and the deposition metrics would be calculated only from sites within the ecoregions. The  
4  $DL_{\%ECO}$  values calculated for each ecoregion (level 3) are presented in Table 5.14s. The  
5  $DL_{\%ECO}$  s calculated for the sensitive category of waterbodies (the CLs from all ecoregions  
6 considered sensitive are pooled together) are compared against the  $DL_{\%ECO}$  developed for each  
7 ecoregions within the sensitivity category for both ANC 50  $\mu\text{eq/L}$  and ANC 20  $\mu\text{eq/L}$  and for  
8 each percentile (90, 75 and 50).

9

10           [Placeholder for additional discussion on percentile values]

11

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10



1 Aquatic and terrestrial critical acid loads were compared in 16 watersheds from each of  
2 the two aquatic acidification Case Study Areas, the Adirondacks and the Shenandoah, identified  
3 in the REA (2009). For each Case Study Area, four watersheds were randomly selected from  
4 each of the four aquatic acidification sensitivity classes reported in the REA (2009). Those four  
5 sensitivity classes are “highly sensitive”, “moderately sensitive”, “low sensitivity”, and “not  
6 sensitive”. In order for a watershed to be classified as one of these four classes, it had to contain  
7 at least one lake or stream with that sensitivity class designation. The Adirondacks Case Study  
8 Area contained watersheds representing all four sensitivity classes, and the 16 watersheds that  
9 were selected for the analysis contained a total of 29 lakes. However, in the Shenandoah Case  
10 Study Area, there were a limited number of watersheds in the “low” and “not sensitive” classes.  
11 Therefore, only one of the 16 randomly selected watersheds contained a “low” and a “not  
12 sensitive” stream. In total, there were 20 streams located in the 16 Shenandoah watersheds  
13 selected for the comparative analysis. In each of the 32 watersheds (16 Adirondacks plus 16  
14 Shenandoah), the terrestrial critical acid loads were calculated as a single value for the entire  
15 watershed. These terrestrial critical acid loads were then compared to the aquatic critical acid  
16 loads for the lakes and streams within each watershed to determine whether the aquatic or  
17 terrestrial critical acid load provided greater protection against acidifying nitrogen and sulfur  
18 deposition. Appendix B of this Policy Assessment document provides a full description of the  
19 methods and results of this comparative analysis.

20 Results of the comparison between the aquatic critical acid load ( $ANC = 50 \mu\text{eq/L}$ ) and  
21 the terrestrial critical acid loads (Bc:Al 1.2 and 10.0) for the 32 watersheds are presented in  
22 **Tables 6.1 and 6.2**. In the 16 Adirondack watersheds, 13 of the 29 lakes had aquatic critical  
23 acid loads that were lower (more protective) than the terrestrial critical acid loads when a Bc:Al  
24 ratio of 10.0 was used. Based on terrestrial critical acid loads determined with a Bc:Al ratio of  
25 1.2, 21 of the 29 lakes in the Adirondacks had aquatic critical acid loads lower than the terrestrial  
26 critical acid loads. More importantly, for the terrestrial critical acid loads determined with a  
27 Bc:Al ratio of 10.0, 13 of the 16 lakes in the Adirondacks classified as “highly” and  
28 “moderately” sensitive to acidification had aquatic critical acid loads lower than the terrestrial  
29 critical acid loads, and all 16 lakes in these two sensitivity classes had critical acid loads lower  
30 than the terrestrial loads determined with a Bc:Al of 1.2. The watersheds within the Shenandoah  
31 region showed similar results (Table 6.1).

1 **Table 6-1. Results of the comparison of lake and stream aquatic critical loads (ANC of 50**  
 2 **µeq/L) to terrestrial critical loads (Bc:Al molar ratios of 10.0 in soil solution) calculated for**  
 3 **the full watershed in each of the 16 watersheds in the Adirondacks and Shenandoah Case**  
 4 **Study Areas.** The tabular results show the number of times the aquatic acidification critical load  
 5 would provide more protection than the terrestrial acidification critical load.

Case Study Area	Watershed Sensitivity to Aquatic Acidification			
	Highly Sensitive	Moderately Sensitive	Low Sensitivity	Not Sensitive
Adirondacks	7 of 7	6 of 9	0 of 7	0 of 6
Shenandoah	14 of 14	5 of 5	0 of 1	0 of 1

6

7 **Table 6-2. Results of the comparison of lake and stream aquatic critical loads (ANC of 50**  
 8 **µeq/L) to terrestrial critical loads (Bc:Al molar ratios of 1.2 in soil solution) calculated for**  
 9 **the full watershed in each of the 16 watersheds in the Adirondacks and Shenandoah Case**  
 10 **Study Areas.** The tabular results show the number of times the aquatic acidification critical load  
 11 would provide more protection than the terrestrial acidification critical load.

Case Study Area	Watershed Sensitivity to Aquatic Acidification			
	Highly Sensitive	Moderately Sensitive	Low Sensitivity	Not Sensitive
Adirondacks	7 of 7	9 of 9	5 of 7	0 of 6
Shenandoah	14 of 14	5 of 5	0 of 1	0 of 1

12

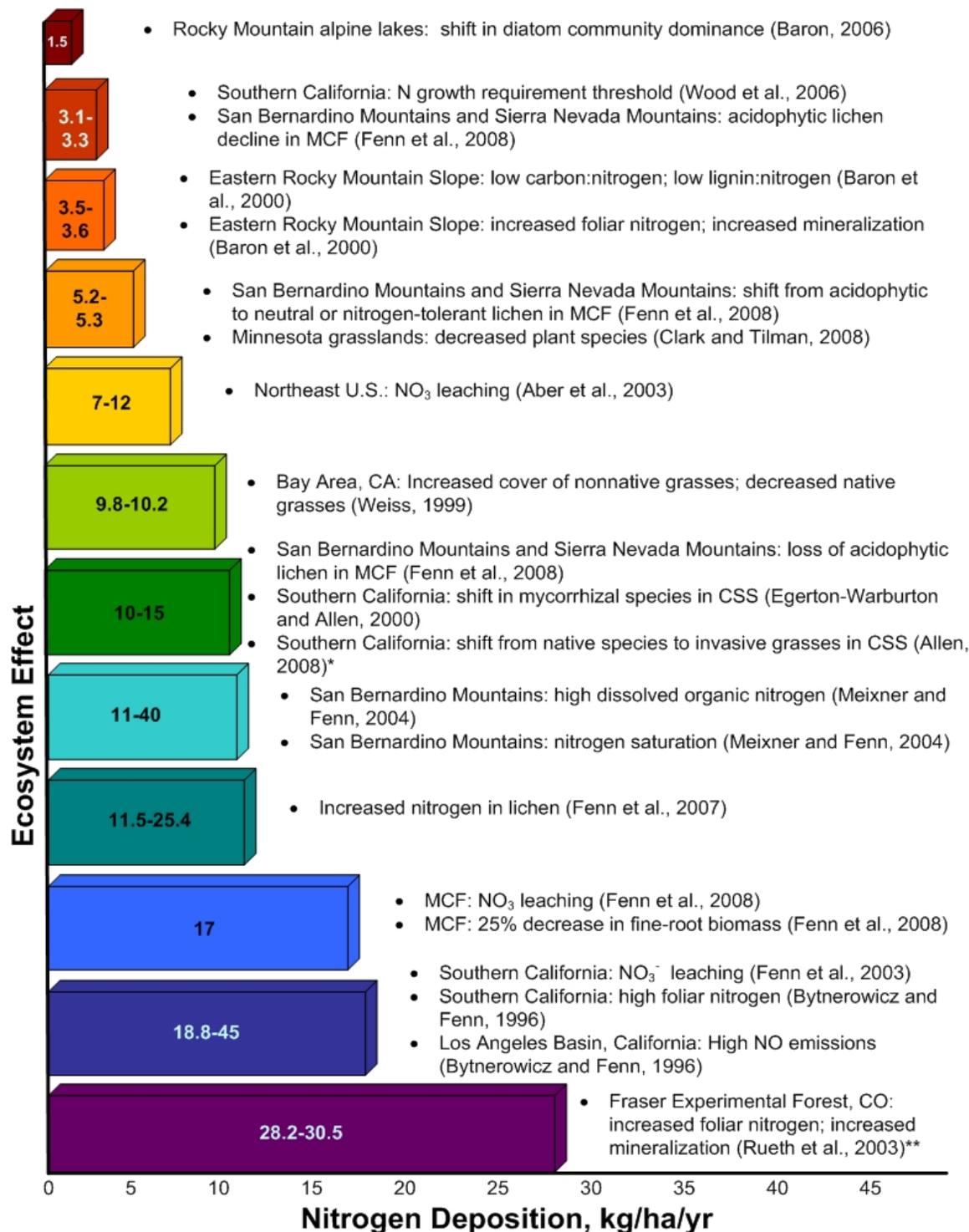
13 In summary, terrestrial and aquatic critical acid loads were compared for watersheds in  
 14 the Adirondack and Shenandoah Case Study Areas. Results indicated that, in general, the  
 15 aquatic critical acid loads were lower and therefore offered greater protection to the watershed  
 16 than did the terrestrial critical acid loads. In situations where the terrestrial critical acid loads  
 17 were lower (i.e., more protective) than the aquatic critical acid loads, the lakes or streams in the  
 18 watershed were often rated as having “low sensitivity” or “not sensitive” to acidifying nitrogen  
 19 and sulfur deposition. Conversely, when the waterbodies were more sensitive to deposition  
 20 (“highly sensitive” or “moderately sensitive”), the aquatic critical acid loads generally provided a

1 greater level of protection against acidifying nitrogen and sulfur deposition in the watershed. It  
2 is uncertain whether these results would be consistent for the rest of the country.

3 **6.2 To What Extent Would A Standard Specifically Defined To Protect Against Aquatic**  
4 **Acidification Likely Provide Protection From Terrestrial Nutrient Enrichment?**

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6 The figure below summarizes the terrestrial nutrient enrichment effects from nitrogen  
7 discussed in the REA (2009). The REA reported these benchmarks as kg/ha/yr. To convert to  
8 meq/m<sup>2</sup>/yr each benchmark number must be multiplied by 7.14. The range in meq/m<sup>2</sup>/yr thus  
9 becomes 10.7 meq/m<sup>2</sup>/yr for the low end benchmark of changes to diatom community structure  
10 to 321 meq/m<sup>2</sup>/yr at the high end benchmark associated with nitrate leaching, high foliar  
11 nitrogen, and high NO emissions in southern California.

12 For each depositional load that is considered for aquatic acidification, whether it is a  
13 national number or a sensitivity based number, it can be compared against the chart in figure 6.1  
14 to understand the level of protection offered in individual parts of the country where these  
15 studies were conducted. For example referring back to the figure under option 1 in section  
16 5.3.2, the depositional load selected to encompass 90% of the critical loads on a national basis to  
17 protect for an ANC of 50 is described as a tradeoff curve between sulfur and nitrogen. If sulfur  
18 were zero then the maximum nitrogen deposition would be 79 meq/m<sup>2</sup>/yr or 11 kg/ha/yr.  
19 Comparing this maximum nitrogen deposition number to the benchmarks in figure 6.1 shows  
20 that the depositional load would provide some protection against leaching in northeast forests,  
21 but would have to be lower to protect California coastal sage scrub, lichens in mixed conifer  
22 forests, alpine lake communities, and Minnesota grasslands.



\* Personal communication, 2008. Also referenced in Bobbink et. al., 2010, Ecological Applications, 20(1):30-59 and USDS FS, 2010, [http://www.nrs.fs.fed.us/clean\\_air\\_water/clean\\_water/critical\\_loads/local-resources/docs/Empirical\\_CLS\\_of\\_N\\_100414.pdf](http://www.nrs.fs.fed.us/clean_air_water/clean_water/critical_loads/local-resources/docs/Empirical_CLS_of_N_100414.pdf)  
 \*\*Nitrogen deposition levels include ambient and experimental additions.

1  
 2 **Figure 6-1 Benchmarks of atmospheric nitrogen deposition for several ecosystem**  
 3 **indicators.**

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**6.3 To What Extent Would A Standard Specifically Defined To Protect Against Aquatic Acidification Likely Provide Protection From Aquatic Nutrient Enrichment?**

The REA (2009) found that deposition of reactive nitrogen contributed to eutrophication of estuaries; however, it was also noted that atmospheric deposition of nitrogen is only part of the total nitrogen load to the estuaries. Due to the complications of separating out the effects of atmospheric deposition from the effects of other nitrogen loads, CASAC did not recommend that a secondary NAAQS be set to specifically protect against estuarine eutrophication at this time.

As described in the REA (2009), the Chesapeake Bay is one national estuary that is suffering from eutrophication. In issuing his Executive Order on the Chesapeake Bay, President Obama recognized that the Bay watershed is one of our nation’s greatest treasures and must be protected and restored. To that end, EPA is proposing a nitrogen total maximum daily load (TMDL) for the Chesapeake Bay. The TMDL will contain a specific air allocation for nitrogen deposition. The allocations that were provided to the states included assumptions that air deposition levels of nitrogen would be reduced to 14.9 million pounds per year to the tidal waters and to 323 million pounds to the watershed by the year 2020. According to the Chesapeake Bay Program Office, the tidal waters have a surface area of 4,479 square miles and the watershed is 64,216 square miles. This means that in 2020, the TMDL currently calls for nitrogen deposition levels to the combined bay and watershed to be reduced to 337.9 million pounds/68,695 square miles/yr, which is equivalent to 8.6 kg/ha/yr or 61 meq/m<sup>2</sup>/yr. As in Section 6.2, this number can be compared to the maximum depositional load of 79 meq/m<sup>2</sup>/yr. If sulfur were zero on the national tradeoff curve from the figure under option 1 of section 5.3.2, then the allowed depositional load of 79 meq/m<sup>2</sup>/yr would not meet the Chesapeake Bay TMDL as currently envisioned.

1 **6.4 REFERENCE**

2 US EPA (United States Environmental Protection Agency). 2009. *Risk and Exposure Assessment*  
3 *for Review of the Secondary National Ambient Air Quality Standards for Oxides of*  
4 *Nitrogen and Sulfur*. Final. U.S. Environmental Protection Agency, Office of Research  
5 and Development, National Center for Environmental Assessment, Research Triangle  
6 Park, NC. September.

7

1       **7 EVALUATION OF UNCERTAINTY AND VARIABILITY IN THE**  
2           **CONTEXT OF AN AAPI STANDARD, INCLUDING MODEL**  
3           **EVALUATION, SENSITIVITY ANALYSES, AND ASSESSMENT OF**  
4           **INFORMATION GAPS**  
5

6       **7.1 INTRODUCTION AND PURPOSE**

7           This chapter provides discussions of results of analyses and assessments intended to  
8 address the relative confidence associated with many of the individual and combined  
9 components of the linked atmospheric-ecological effects system described in Chapter 5, as well  
10 as important uncertainties in the scientific evidence that should be considered in developing  
11 options for the standard. This chapter is intended to integrate a variety of analyses related to the  
12 sensitivity of the models and model components to uncertainty and variability, and place the  
13 results of those analyses within the context of the conclusions that can be drawn regarding the  
14 components of the AAPI. These components include ecosystem effects; dose-response  
15 relationships; underlying ecosystem sensitivity to acid deposition, biogeochemical, atmospheric  
16 and deposition processes; and characterization of ecosystem services. While several processes  
17 are imbedded in the AAPI equation introduced in Chapter 5, the level of the AAPI, as in all  
18 NAAQS, is to include consideration of information on uncertainty and variability.  
19 Consequently, knowledge of the relative confidence and natural variability in the structural  
20 components of the AAPI are considered in staff conclusions on options for ranges of the level of  
21 the standard. This chapter is not intended to be a comprehensive treatment of all uncertainties  
22 that exist relative to the overall review of the standards, instead, it focused on those that are most  
23 relevant in evaluating choices regarding the AAPI form of the standard and options regarding the  
24 indicator, averaging time, and ranges of levels of the AAPI-based NO<sub>x</sub> and SO<sub>x</sub> standard.

25           Uncertainty and sensitivity analyses are used to inform the relative confidence in the  
26 components and models that are used in defining the standard. Assessments of variability in the  
27 data used to determine parameters of the standard increases the level of understanding about the  
28 likelihood that alternative parameterizations of the standard will achieve targeted levels of  
29 protection when applied to sensitive ecosystems across the U.S. Assessments of the sensitivity  
30 of the overall AAPI to the components of the equation proposed to calculate the AAPI can help

1 demonstrate how important uncertainty and variability in those components are in assessing the  
2 protection of ecosystems provided by an AAPI standard. To evaluate the potential interactions  
3 between uncertain and/or variable AAPI components, a multifactor sensitivity analysis is also  
4 conducted. The ranges of component values evaluated in the multifactor sensitivity assessment  
5 are guided by individual variability and uncertainty analyses of specific components. In addition  
6 to informing considerations of the AAPI level, an additional objective of these “confidence”  
7 related analyses and discussions is to help guide research and data collection efforts intended to  
8 reduce uncertainty for future NAAQS reviews and implementation efforts. Spatial and  
9 temporal variability analyses of AAPI components are especially useful to inform monitoring  
10 network design, the spatial boundaries of acid sensitive regions, and averaging periods relevant  
11 to NAAQS implementation.

12 *Use of confidence assessment results to inform setting of the AAPI level.*

13 The analyses summarized in this chapter largely address parameters related to  
14 atmospheric/deposition and biogeochemical characterization, which are incorporated in the  
15 AAPI. General uncertainty related discussions are provided for elements in the standard setting  
16 process including dose-response relationships, effects, economic valuation metrics, as well as for  
17 several specific processes (e.g., organic nitrogen) which are directly represented in the AAPI.  
18 Confidence related information is not used to make decisions on inclusion or exclusion of a  
19 parameter in the AAPI model. Instead, we develop the overall approach using the best available  
20 information and most practical approaches, with an understanding of the level of confidence  
21 associated with both individual components and the overall resulting AAPI. The overarching  
22 principle in setting the level of the AAPI is consideration of the impact that associated NO<sub>x</sub> and  
23 SO<sub>x</sub> levels will have on the level of protection from adverse effects on public welfare.  
24 Information on uncertainty in different elements of the form of the standard and the scientific  
25 evidence informing those elements is used to evaluate our confidence that the standard has a high  
26 likelihood of protecting public welfare. One way of considering confidence levels is to  
27 identify components of the standard for which we have low confidence and adjust the level of the  
28 AAPI to ensure that, given the uncertainty in those components, we have a high likelihood of  
29 achieving a target level of protection. This approach to considering confidence levels places  
30 greater weight on ensuring a high likelihood of achieving protection, and places less weight on

1 concerns about providing more than requisite protection. Conversely, low confidence in  
2 components of the standard could also be considered to increase the likelihood that more than  
3 requisite protection could result, and consequently, the AAPI could be adjusted downward to  
4 account for lower confidence that the level of protection is requisite. To the extent that the  
5 available information suggests that a particular process or parameter creates a positive or  
6 negative bias (as opposed to broader uncertainty with no clear direction) would lead to  
7 recommending correspondingly higher or lower AAPI level. The Clean Air Act (CAA)  
8 language requires the secondary standards to be set to provide a requisite level of protection  
9 against known or anticipated effects, which can be interpreted to include effects which have  
10 large uncertainty but for which the expectation of adverse effects on public welfare exists with  
11 reasonable confidence.

12 Significant emphasis is placed on evaluations of CMAQ due to the unique role that  
13 atmospheric models hold in the linked AAPI system. The AAPI as currently formulated relies  
14 on CMAQ for both the initial characterization of reduced nitrogen deposition, and the deposition  
15 transformation ratios ( $T_{NOx}$  and  $T_{SOx}$ ) which characterize the relationships between atmospheric  
16 concentrations of  $NOx$  and  $SOx$  and deposition of N and S. Included are interpretations of  
17 model evaluation results from the REA (EPA, 2009) as well as more recent results related to wet  
18 deposition and the treatment of ammonia deposition. Comparison of model results to  
19 observations provides a general sense of the confidence we have that the models capture the  
20 spatial, temporal and compositional texture of the relevant atmospheric and deposition species  
21 that drive the linked atmospheric-ecosystem processes. Both model evaluation results and  
22 assessments of spatial and temporal variability guide implementation strategies for monitoring  
23 network design and emission inventory improvement. Sensitivity of CMAQ derived deposition  
24 transformation ratios to changes in emissions, and treatment of chemistry [not yet completed]  
25 and variability over time provide insight into the stability of these parameters that are used in a  
26 relatively static manner in the AAPI, and into how well proposed averaging times capture the  
27 overall spatial and temporal trends in the parameters.

28 We evaluate the sensitivity of critical load modeling components by comparing dynamic  
29 (MAGIC) and hybrid steady state model results, looking at terminal results of MAGIC. This  
30 approach was viewed as a test of the more reduced form approximations used in steady state

1 modeling relative to more sophisticated treatment in MAGIC. The MAGIC critical load  
2 simulations also provide information on the temporal trajectory of ANC, including the expected  
3 time necessary to reach a desired ANC, which can help inform the level of the AAPI,  
4 recognizing that there may be additional consideration given to reaching a target ANC within a  
5 specific timeframe, e.g. by 2030 or 2040. [[not yet completed]]

6 For the purposes of this discussion, we characterize *uncertainty* regarding models and  
7 their outputs as referring to the lack of knowledge regarding both the actual values of model  
8 input variables (parameter uncertainty) and the model characterization of physical systems or  
9 relationships (model uncertainty). In any application, uncertainty is, ideally, reduced to the  
10 maximum extent possible, but significant uncertainty often remains. It can be reduced by  
11 improved measurement and improved model formulation. Model evaluation results provide  
12 some insight into the relative uncertainty associated with the ability of models to capture key  
13 environmental state characteristics. Confidence regarding the fundamental science supporting  
14 causal determinations about the effects of acid deposition, and the translation of those effects  
15 into ecosystem services and values is less amenable to quantification. As a result, these  
16 uncertainties are more difficult to explicitly account for in development of the standards. In the  
17 case of the equation describing the AAPI, while the degree of uncertainty in some elements can  
18 be characterized, sometimes quantitatively, a formal uncertainty analysis using statistical  
19 sampling techniques (e.g., Monte Carlo simulation) to identify the relative and combined  
20 influences of parameter uncertainty was not performed. However, we did evaluate the sensitivity  
21 of the AAPI to its components using two related assessments: analysis of variance (ANOVA)  
22 and elasticity calculations. The results of these assessments are addressed in section 7.5.

23  
24 *Sensitivity* refers to the influence on modeled results due to perturbations in input  
25 variables or change of process formulations. Sensitivity analysis can provide a sense of how  
26 important different parameters and inputs might be to the outcomes of interest, e.g. the AAPI  
27 level, but cannot by themselves indicate how important specific parameters actually are, because  
28 they do not incorporate information on the range of parameter values or the likelihood associated  
29 with any specific parameter value. Sensitivity results in this PAD are intended to provide  
30 insight into the relative stability of the AAPI and associated NO<sub>x</sub> and SO<sub>x</sub> tradeoff curves and  
31 confidence in modeled parameterizations. Sensitivity analyses are especially useful in the

1 absence of observed data to challenge models. For example, the NO<sub>y</sub> and SO<sub>x</sub> transference  
2 ratios are a model construct that is difficult, if not impossible, to compare to observations. The  
3 sensitivity of these ratios to changing meteorology, emissions and chemical mechanism  
4 treatments is evaluated in reference to the stability of these ratios under changing conditions.  
5 Low sensitivity here implies that the choice to use long-term averages of modeled ratios is  
6 justified. Sensitivity analyses also are used to discern the relative influence (on AAPI results) of  
7 AAPI parameters. Toward that end, ANOVA and elasticity analyses were applied to determine  
8 the relative sensitivity of AAPI results associated with individual and combined AAPI  
9 parameters.

10 *Variability* refers to the heterogeneity in a population or variable of interest that is  
11 inherent and cannot be reduced through further data collection and research. In the context of  
12 the AAPI and trade-off curves, variability is considered in guiding the design of monitoring and  
13 modeling analyses supporting implementation activities.

14

## 15 **7.2 Uncertainty associated with ecosystem effects and dose – response relationships.**

16 Chapter 2 provides a brief summary of uncertainties based on the REA and is reproduced  
17 here to centralize all uncertainty discussions. There are different levels of uncertainty associated  
18 with relationships between deposition, ecological effects and ecological indicators. In Chapter 7  
19 of the REA, the case study analyses associated with each targeted effect area were synthesized  
20 by identifying the strengths, limitations, and uncertainties associated with the available data,  
21 modeling approach, and relationship between the ANC and atmospheric deposition. The key  
22 uncertainties were characterized as follows to evaluate the strength of the scientific basis for  
23 setting a national standard to protect against a given effect (**REA 7.0**):

- 24 • **Data Availability: *high, medium or low quality*.** This criterion is based on the  
25 availability and robustness of data sets, monitoring networks, availability of data that  
26 allows for extrapolation to larger assessment areas, and input parameters for modeling  
27 and developing the ecological effect function. The scientific basis for the ecological  
28 indicator selected is also incorporated into this criterion.
- 29 • **Modeling Approach: *high, fairly high, intermediate, or low confidence*.** This value is  
30 based on the strengths and limitations of the models used in the analysis and how  
31 accepted they are by the scientific community for their application in this analysis.

- 1 • **Ecological Effect Function: *high, fairly high, intermediate, or low confidence.*** This  
2 ranking is based on how well the ecological effect function describes the relationship  
3 between atmospheric deposition and the ecological indicator of an effect.  
4

5 The REA concludes that the available data are robust and considered *high quality*. There is  
6 high confidence about the use of these data and their value for extrapolating to a larger regional  
7 population of lakes. The EPA TIME/LTM network represents a source of long-term,  
8 representative sampling. Data on sulfate concentrations, nitrate concentrations and ANC from  
9 1990 to 2006 used for this analysis as well as EPA EMAP and REMAP surveys, provide  
10 considerable data on surface water trends.

11 There is *fairly high confidence* associated with modeling and input parameters. Uncertainties  
12 in water quality estimates (i.e., ANC) from MAGIC was derived from multiple site calibrations.  
13 The 95% confidence interval for pre-acidification of lakes was an average of 15 µeq/L difference  
14 in ANC concentrations or 10% and 8 µeq/L or 5% for streams (**REA 7.1.2**). The use of the  
15 critical load model used to estimate aquatic critical loads is limited by the uncertainties  
16 associated with runoff and surface water measurements and in estimating the catchment supply  
17 of base cations from the weathering of bedrock and soils (McNulty et al., 2007). To propagate  
18 uncertainty in the model parameters, Monte Carlo methods were employed to develop an inverse  
19 function of exceedences. There is *high confidence* associated with the ecological effect function  
20 developed for aquatic acidification. In calculating the ANC function, the depositional load for N  
21 or S is fixed by the deposition of the other, so deposition for either will never be zero (**Figure**  
22 **7.1-6 REA**).

23 Chapter 2 also reviews the basic evidence underlying effects on fish mortality, aquatic species  
24 diversity and more extended food web disruptions leading to adverse impacts on birds associated  
25 with aquatic acidification. There is high confidence associated with causality between  
26 acidification and these ecological effects. Also, there is extremely high confidence in the  
27 relationship between the ecological indicator, ANC, and the more direct chemical properties  
28 (lower pH and increased Al) associated with acidification.  
29  
30

### 1   **7.3    Uncertainty in benefits estimates**

2

3           Descriptions of the current provision of ecosystem services presented for each of the  
4 effect areas analyzed for this review followed by estimations of the damages incurred to selected  
5 services due to nitrogen and sulfur deposition. The current services are presented to give the  
6 reader a sense of the magnitude of the benefit the public receives from these ecosystems under  
7 current conditions. The data used in these descriptive passages is generally derived from  
8 government (either federal or state) sources we are reasonably certain to be of the highest  
9 quality. Where monetary values are placed on these services we have generally used widely  
10 cited studies, particularly meta analyses that provide an average value that smoothes the variation  
11 in WTP estimates. These estimates underestimate the total value of these services as they use  
12 benefit estimates for a marginal increase in these services. It is likely that the total benefits of  
13 these services are greater because their marginal value likely is lower than the average value.  
14 While reductions in sulfur and nitrogen emissions would increase the size of the benefits from  
15 these services, for many of them it is unknown how significant the increase will be.

16           The analyses of damages incurred are more uncertain and are limited to those areas where  
17 data and tools were available. Only some services were analyzed which in some cases meant  
18 that the results were limited to one or two services and in the case of terrestrial nutrient  
19 enrichment no services had sufficient data available to attempt an estimate of damage. This  
20 means that the estimates presented are a very small part of the total damage incurred due to  
21 deposition.

#### 22    *Aquatic Acidification*

#### 23    **Recreational Fishing Model.**

24           The analysis of recreational fishing damages presented in Chapter 3 is subject to the  
25 assumptions necessary to perform the analysis. The original analysis performed for the REA  
26 was based on projecting future benefits of increased recreational fishing based on a complete  
27 cessation of all nitrogen and sulfur emissions. These decisions under or over estimate the current  
28 damages to public welfare incurred from nitrogen and sulfur deposition. The magnitude of the  
29 bias in results is unknown in either direction however the majority of the assumptions influence

1 the estimates downward. These include the use of emissions estimates that include projected  
2 decreases due to implementation of Title IV regulations in 2020. These emissions estimates are  
3 lower than current emissions and therefore lead to underestimation of damages. Because the  
4 models only value this improvement for New York residents (without accounting for out-of-state  
5 visitors) the damages are underestimates of the benefits of these improvements in the  
6 Adirondacks region.

7 The use of projected population in the REA analyses contributes to an overestimate of  
8 current damages since current population is smaller than future population. Further, these  
9 estimates are extrapolated from a 44 lake subset and applied to all Adirondack lakes. The  
10 representativeness of this sample is unknown. This analysis also does not account for any  
11 change in fishing demand (possible overestimate) and income (possible underestimate).

#### 12 **Banzhaf, et al Benefits Transfer.**

13 The approach using the WTP estimates from the Banzhaf study is subject to the same  
14 uncertainties described above and some additional considerations. Specifically there is some  
15 uncertainty regarding which types of ecosystem services are reflected in the study's estimates of  
16 the improvements in ecosystem services of reducing acidification, particularly provisioning and  
17 regulating services. The values likely include recreational fishing services, which means *they*  
18 *cannot be added to the recreational fishing model results*, and other cultural services including  
19 other recreation and nonuse services. The inclusion in the survey of other ecosystem changes  
20 (birds, trees, etc.) leads to an overestimation of WTP for remediation of lake acidification alone.  
21 Finally, assumptions were required to align the Banzhaf survey scenarios to the likely results of  
22 complete removal of all nitrogen and sulfur emissions. These are reasonably close but not exact  
23 and may not be applicable to another baseline.

#### 24 **Conclusion.**

25 While these estimates are subject to uncertainty we are reasonably confident that they  
26 represent a good first-order approximation of the damages to recreational fishing due to nitrogen  
27 and sulfur deposition. Additionally it should be noted that the Banzhaf survey results represent a  
28 broader picture (though by no means complete) of the damages to ecosystem services in the  
29 Adirondacks. Finally, we would again like to emphasize that these estimates represent only a

1 small sample of the damages incurred to a broad range of ecosystem services affected and the  
2 areas of the nation where acidic deposition is an ongoing issue.

### 3 **7.4 CMAQ Application and Evaluation**

#### 4 **7.4.1 Overview of CMAQ model application**

5 The CMAQ model is a comprehensive, peer-reviewed (Aiyyer et al., 2007), three-  
6 dimensional grid-based Eulerian air quality model designed to simulate the formation and fate of  
7 gaseous and particle (i.e., particulate matter or PM) species, including ozone, oxidant precursors,  
8 and primary and secondary PM concentrations and deposition over urban, regional, and larger  
9 spatial scales (Dennis et al., 1996; U.S. EPA, 1999; Bryun and Schere, 2006). CMAQ is run for  
10 user-defined input sets of meteorological conditions and emissions. For this analysis, we are  
11 using predictions from several existing CMAQ runs. These runs include annual simulations for  
12 2002 using CMAQv4.6 and annual simulations for each of the years 2002 through 2005 using  
13 CMAQv4.7 (Foley et al., 2010). CMAQv4.6 was released by the U.S. Environmental Protection  
14 Agency's (EPA's) Office of Research and Development (ORD) in October 2007. CMAQv4.7  
15 along with an updated version of CMAQ's meteorological preprocessor (MCIPv3.4, Otte and  
16 Pleim, 2010)<sup>1</sup> were released in October 2008<sup>2</sup>. The 2002 simulation with CMAQv4.6 was  
17 performed for both the Eastern and Western domains. The horizontal spatial resolution of the  
18 CMAQ grid cells in these domains is 12 x 12 km. The 2002 through 2005 simulations with  
19 CMAQv4.7 were performed for the eastern 12-km domain and for the continental United States  
20 domain, which has a grid resolution of 36 x 36 km. The CMAQv4.6 and v4.7 annual simulations  
21 feature year-specific meteorology, as well as year-specific emissions inventories for key source  
22 sectors, such as utilities, on-road vehicles, nonroad vehicles, wild fires, and natural biogenic  
23 sources. Emissions for other sectors of the inventory for each of the years modeled rely on  
24 inventories for 2002. Details on the development of emissions, meteorology, and other inputs to  
25 the 2002 CMAQv4.6 runs can be found in a separate report (U.S. EPA, 2008). Inputs for the  
26 CMAQv4.7 runs for 2002 through 2005 were derived using procedures similar to those for the  
27 CMAQv4.6 2002 runs.

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<sup>1</sup> The scientific updates in CMAQ v4.7 and MCIP v3.4 can be found at the following web links:  
[http://www.cmascenter.org/help/model\\_docs/cmaq/4.7/RELEASE\\_NOTES.txt](http://www.cmascenter.org/help/model_docs/cmaq/4.7/RELEASE_NOTES.txt)  
[http://www.cmascenter.org/help/model\\_docs/mcip/3.4/ReleaseNotes](http://www.cmascenter.org/help/model_docs/mcip/3.4/ReleaseNotes)

<sup>2</sup> The differences in nitrogen and sulfur deposition in the case study areas between CMAQ v4.6 and v4.7 for 2002 are small, as described in Chapter 3.

1 Additional details of the modeling domain, emissions and meteorological inputs are  
2 provided in EPA (2009; REA Appendices).

#### 3 4 **7.4.2 CMAQ Evaluation, Sensitivity and Variability Analyses**

5 **Past results**. A variety of comparisons of modeled estimates to observations were  
6 included in the REA (EPA, 2009), and some of the highlights are summarized here in addition to  
7 new work on ammonia characterization and wet deposition. Readers are encouraged to review  
8 the earlier report. Ambient air concentrations and wet deposition observations are paired against  
9 modeled estimates. In contrast, dry deposition is always a modeled value, either derived from  
10 ambient or modeled ambient concentrations. Given the interest in relevant nitrogen and sulfur  
11 species, CASTNET observations were used extensively. Comparisons of modeled annual  
12 average total nitrate (sum of nitric acid and particulate nitrate), ammonium, sulfate, and sulfur  
13 dioxide to observations for the 2002 base year are provided in Figures 7-1 through 7-4.  
14 Normalized mean bias statistics for 2002-2005 base years are provided in Table 7-1.

15 CMAQ overpredicts SO<sub>2</sub> and underpredicts SO<sub>4</sub>. Although model performance is good  
16 for total SO<sub>x</sub>, the inclusion of co-located SO<sub>2</sub> and sulfate measurements required for future  
17 secondary NO<sub>x</sub>/SO<sub>x</sub> NAAQS comparisons will help diagnose issues with the model's ability to  
18 partition these two species. CMAQ generally overpredicts total nitrate and slightly  
19 underpredicts ammonium and the model captures the monthly temporal patterns of sulfate, total  
20 nitrate and ammonium when all sites are aggregated (Figures 7-5 to 7-7). There are some basic  
21 incommensurabilities between model estimates and observations that complicate interpretation  
22 of model to observation comparisons, most notably the representation of space as a model  
23 represents a volume average of roughly 144 km<sup>2</sup>, which depends on the time varying vertical  
24 depth of the lowest modeled layer. Most surface based observations rely on point sampling and  
25 the extent to which a point is representative of broader volume space varies with meteorology,  
26 distribution of emissions and surface characteristics.

27

1 **Table 7-1. Normalized Mean Bias Statistics for Predicted and Observed Pollutant**  
 2 **Concentration**

<b>Pollutant Concentrations</b>	<b>2002</b>	<b>2003</b>	<b>2004</b>	<b>2005</b>
SO <sub>2</sub>	45%	39%	47%	41%
SO <sub>4</sub> <sup>2-</sup>	-13%	-9%	-13%	-17%
TNO <sub>3</sub>	22%	26%	22%	24%
NH <sub>4</sub> <sup>+</sup>	4%	11%	7%	2%

3 **Ammonia.**

4 Characterizing ammonia deposition is of increasing importance for assessing ecosystem  
 5 responses to nitrogen deposition. Nitrogen deposition includes both deposition of NO<sub>y</sub> and  
 6 deposition of reduced nitrogen (ammonia + ammonium). Ecosystem effects are due to total  
 7 nitrogen and, as such, the recommended AAPI form of the standard directly includes reduced  
 8 nitrogen to account for its role in using up an ecosystem's ability to absorb nitrogen. The  
 9 proposed form of the standard uses CMAQ modeled deposition of reduced nitrogen.

10 Because of a shortage of routinely available ammonia observations, model evaluation  
 11 studies have relied on ammonium measurements from CASTNET as the only routine source of  
 12 reduced nitrogen observations. Clearly, the lack of ammonia observations must be addressed in  
 13 future implementation scenarios as there is relatively greater uncertainty in characterizing  
 14 ammonia relative to SO<sub>x</sub> and NO<sub>y</sub>. Recently, Dennis et al. (2011) explored the sensitivity of  
 15 CMAQ (addressed below) to varying treatments affecting ammonia deposition velocity. As part  
 16 of that study, CMAQ ammonia estimates were compared with ammonia observations at two sites  
 17 in North Carolina (Figure 7-8).

18 **Wet deposition.**

19 Modeled wet deposition in CMAQ is a function of the volume of predicted precipitation  
 20 within a grid cell and the pollutant concentrations scavenged from the atmosphere during  
 21 precipitation events. As a result, errors in modeled precipitation and in emission inputs can lead  
 22 to significant bias and error in the wet deposition predictions compared to observed values. EPA

1 (Dennis and Foley, 2010) has corrected CMAQ wet deposition predictions by scaling the model  
2 output based on observation-based gridded precipitation data generated by the Parameter-  
3 elevation Regressions on Independent Slopes Model (PRISM, 2004). The precipitation adjusted  
4 deposition fields are more highly correlated with observed values for all wet deposited nitrogen  
5 and sulfur species compared to the base model output (Figures 7-9). In addition, the adjusted  
6 fields are better able to capture the spatial heterogeneity of accumulated wet deposition due to  
7 orographic effects on precipitation amounts.

8 Adjusting the wet deposition values to account for over-predictions in the model  
9 precipitation inputs revealed compensating errors for nitrate and ammonium. The negative bias  
10 seen in these species after the precipitation adjustment is believed to be due to missing emissions  
11 sources. A second bias adjustment was performed for nitrate and ammonium based on observed  
12 levels at the NADP/NTN sites (Figure 7-10). The final adjusted spatial fields of annual total wet  
13 deposition values are more consistent with observed wet deposition values. Ongoing studies  
14 suggest that much of this bias can be reduced in the Eastern half of the US by including nitrogen  
15 oxide produced by lightning and accounting for the bi-directional flux of ammonia. Once these  
16 model improvements are incorporated in CMAQ a second bias adjustment may not be needed in  
17 the East.

### 18 **7.4.3 Variability and sensitivity of CMAQ generated components.**

#### 19 **7.4.3.1 Ambient Concentration to Deposition Transformation ratios.**

20 The deposition transference ratios<sup>3</sup> introduced in Chapter 5 are referenced as  $T_{SO_x}$  and  
21  $T_{NO_y}$ , to distinguish these parameters from an exact linkage to deposition velocity, which is  
22 uniquely associated with individual atmospheric species. Deposition transference ratios are  
23 defined as the annual wet and dry deposition of all oxidized species ( $NO_y$  for  $T_{NO_y}$ ,  $SO_2$  plus  $SO_4$   
24 for  $T_{SO_x}$ ) divided by the average annual concentration of  $NO_y$ , for  $T_{NO_y}$ , or  $SO_4$  plus  $SO_2$ , for  
25  $T_{SO_x}$ . The units for  $T_{NO_y}$  and  $T_{SO_x}$  are distance/time. Deposition transference ratios provide a  
26 mechanism to associate ambient concentrations to deposition loads and to determine if an area's

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<sup>3</sup> In the first draft of the Policy Assessment, the deposition transformation ratios were labeled  $V_{NO_x}$  and  $V_{SO_x}$ . For this draft, based on recommendations from CASAC, we have renamed these ratios  $T_{NO_x}$  and  $T_{SO_x}$ .

1 air concentrations of  $\text{NO}_y$  and  $\text{SO}_x$  meet a NAAQS level using the AAPI form. A deposition  
2 transformation ratio is an aggregate representation of the deposition process generated through  
3 modeling which does not lend itself to a traditional analysis relating observations and  
4 predictions. Furthermore, there is an implicit assumption that the response of deposition  
5 transformation ratios to changes in meteorology and emissions is relatively stiff, as these ratios  
6 are an attribute of the system that channels ambient air response associated with decreases in  
7 emissions of  $\text{NO}_x$  and  $\text{SO}_x$  to changes in deposition. The stiffness of the deposition transference  
8 ratios would suggest that the relationship between ambient concentrations and deposition is  
9 strictly a constant proportion, not impacted by the mixture and level of emissions or by changes  
10 in meteorology. To better understand the implications of this assumption, we investigated the  
11 relative variability of the modeled deposition transformation ratios across time and space, and the  
12 stability of the ratios relative to emissions and meteorological inputs was conducted to guide  
13 EPA in determining how uncertainties in this parameter may eventually impact AAPI related  
14 calculations.

#### 15 **7.4.3.2 Spatial and Interannual Variation of $T_S$ and $T_N$ .**

16 Generally small spatial and inter-annual variability exist in the deposition transformation  
17 ratios for the 2002 -2005 model years (Figure 7-11). The inter-annual variability, calculated at  
18 the grid cell level, as measured by the median coefficient of variation is around 10% and the  
19 absolute values of the ratios remain stable (Figure 7-12), suggesting that year to year changes in  
20 meteorology have minimal impact on the ratios. Spatial homogeneity of deposition  
21 transformation ratios within the two acid sensitive areas we evaluated in the REA (Adirondacks  
22 and Shenendoah) (Figure 7-11) is consistent with a relatively homogeneous ambient  
23 concentration environment overlaid upon a landscape of similar vegetation and surface  
24 conditions. Such spatial homogeneity within case study areas provides confidence that an area  
25 wide application AAPI will not be strongly dependent on the exact boundaries chosen to define  
26 an acid sensitive area.

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1 **7.4.3.3 T<sub>SOx</sub> and T<sub>NOy</sub> Sensitivity to emission changes.**

2 The response of T<sub>SOx</sub> and T<sub>NOy</sub> to emission changes was explored by analyzing available  
3 base case 2005 and 2030 CMAQ simulations. The 2030 case reflected expected changes in  
4 emissions associated with simulated implementation of a variety of national rules and represents  
5 Eastern U.S. domain wide NOx and SOx emission reductions of 48% and 40%, respectively.  
6 Median changes in deposition transference ratios tended to be around zero (figure 7-12), with the  
7 Adirondack region exhibiting slightly higher response than the Shenandoah region and  
8 remainder of the Eastern U.S. domain.

9 **7.4.3.4 T<sub>SOx</sub> and T<sub>NOy</sub> sensitivity to different chemical mechanisms.**

10 [Intended to be added in the final PA]

1 **7.4.3.5 Ammonia sensitivity.**

2 The role of  $\text{NH}_x$  deposition is incorporated in the AAPI expression as a parameter that  
3 influences the level of allowable concentrations of  $\text{NO}_y$  and  $\text{SO}_x$ , due to its role as part of the  
4 total reactive nitrogen budget which affects acidification.. Characterizing ammonia deposition  
5 is challenging due to the variety of surface and vegetation types that influence ammonia dry  
6 deposition velocities as well the potential for bi-directional flux of ammonia. In addition,  
7 ammonia emission estimates remain relatively more uncertain than emissions of  $\text{NO}_x$  and  $\text{SO}_2$   
8 given the complexity of meteorology and agricultural practices that influence the spatial and  
9 temporal patterns of ammonia releases. An exploration of the sensitivity of ammonia to three  
10 different treatments of deposition processes in CMAQ was performed by EPA (Dennis et al.,  
11 2010) to test the inclusion of a bi-directional  $\text{NH}_3$  flux algorithm and elucidate the relative  
12 importance associated with advection, deposition and chemical transformation on ammonia  
13 patterns. These treatments included a (1) base case of current CMAQ treatment using existing  
14 ammonia deposition velocity schemes and uni-directional deposition, (2) modified the base case  
15 by replacing ammonia deposition velocity calculations with  $\text{SO}_2$  deposition velocities ( $\text{SO}_2$   
16 interacts with surfaces and vegetation similarly to  $\text{NH}_3$ , but with reduced velocity) as a lower  
17 bound and (3) introducing a bi-directional flux algorithm to the base case (retaining  $\text{NH}_3$   
18 deposition velocities). Based on modeled process analysis that delineates the effects of  
19 deposition, chemical transformation and advection (horizontal and vertical) on emitted ammonia,  
20 the results (Figure 7-13) suggest that ammonia patterns, especially when a bi-directional flux  
21 process is incorporated, are more indicative of a transported pollutant where emissions influence  
22 can span hundreds of kilometers, markedly different from some earlier perspectives where  
23 ammonia often was thought of as near source phenomenon due to high deposition velocities.  
24 The process analysis illustrates the importance of vertical advection which enables the movement  
25 of ammonia into traditional mesoscale flow patterns. The effect is enhanced by the  
26 reintroduction of deposited ammonia through bi-directional flux into the ambient environment.

27 From a monitoring perspective, a design that addresses the regional characterization of  
28  $\text{NO}_y$  and  $\text{SO}_x$  would be consistent with characterizing  $\text{NH}_x$ . Not only would ammonia and  
29 ammonium measurements be useful for estimating dry deposition through deposition modeling  
30 approaches such as those used in CASTNET, but they would serve as important diagnostic data

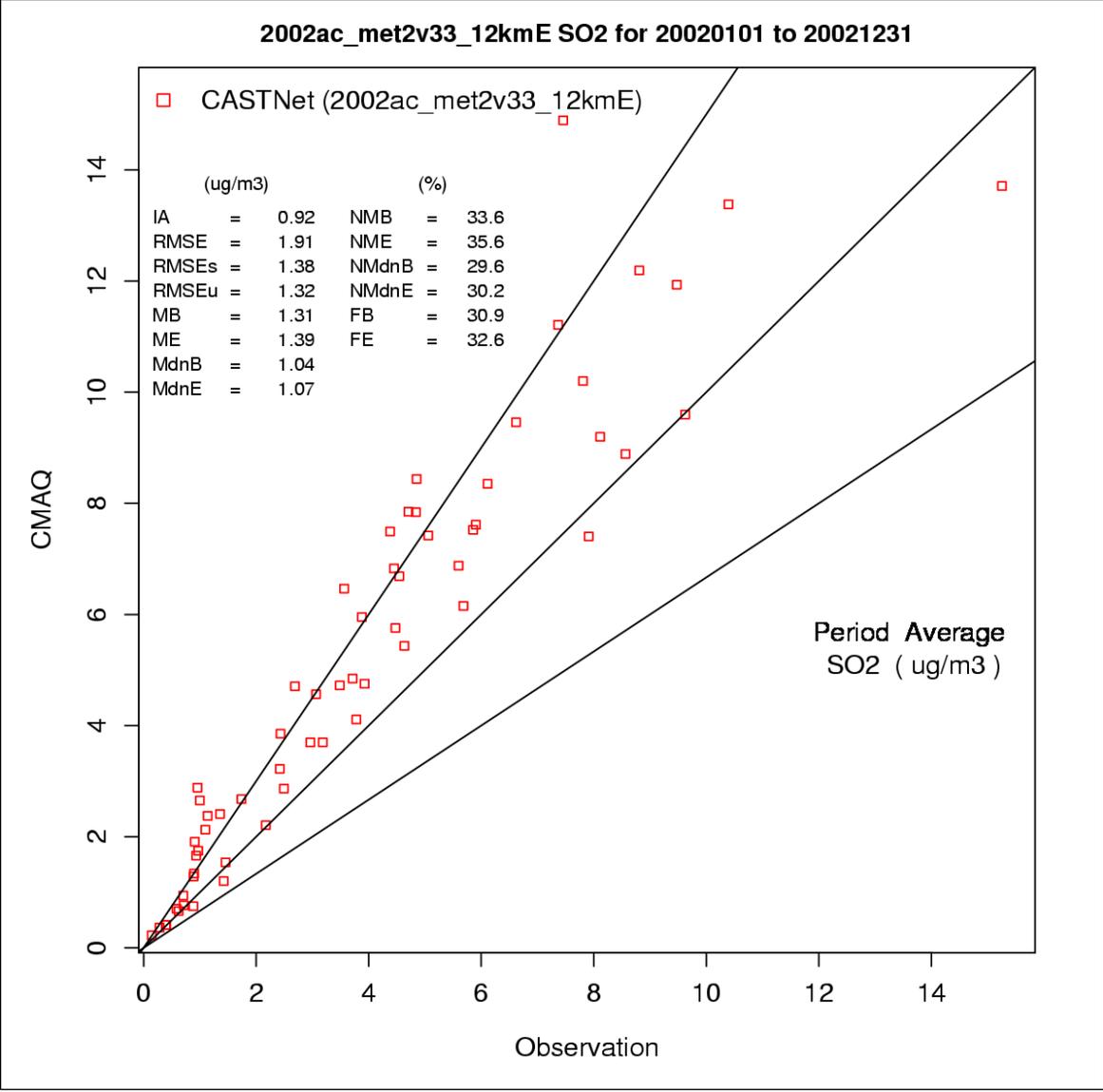
1 to continually assess the effectiveness of  $\text{NH}_x$  deposition processes in models like CMAQ. This  
2 is especially important as we recognize a large uncertainty in the bi-directional formulation  
3 associated with the estimation of  $\Gamma$ , the emissions potential due to the existence of compensation  
4 points. Nonetheless, we can learn much about the  $\text{NH}_3$  budget in spite of these uncertainties.

5 High priority research is ongoing to improve the bi-directional parameterization and the  
6 estimates of the leaf and soil gammas across different cropping regions and throughout the year.  
7 We are developing a software tool to estimate the soil  $\Gamma$  associated with fertilizer application.  
8 When we have a spatially and temporally varying  $\Gamma_g$ , we will investigate the emissions budgets  
9 for fertilized fields, as well as reexamine the animal operation emission budgets, as this will be  
10 of interest. Work to examine the seasonality of single cell budgets and their range of influence is  
11 continuing. Current and future CMAQ applications to ecosystem deposition will incorporate bi-  
12 directional flux treatment of ammonia.

#### 13 **7.4.3.6 CMAQ uncertainties and the AAPI**

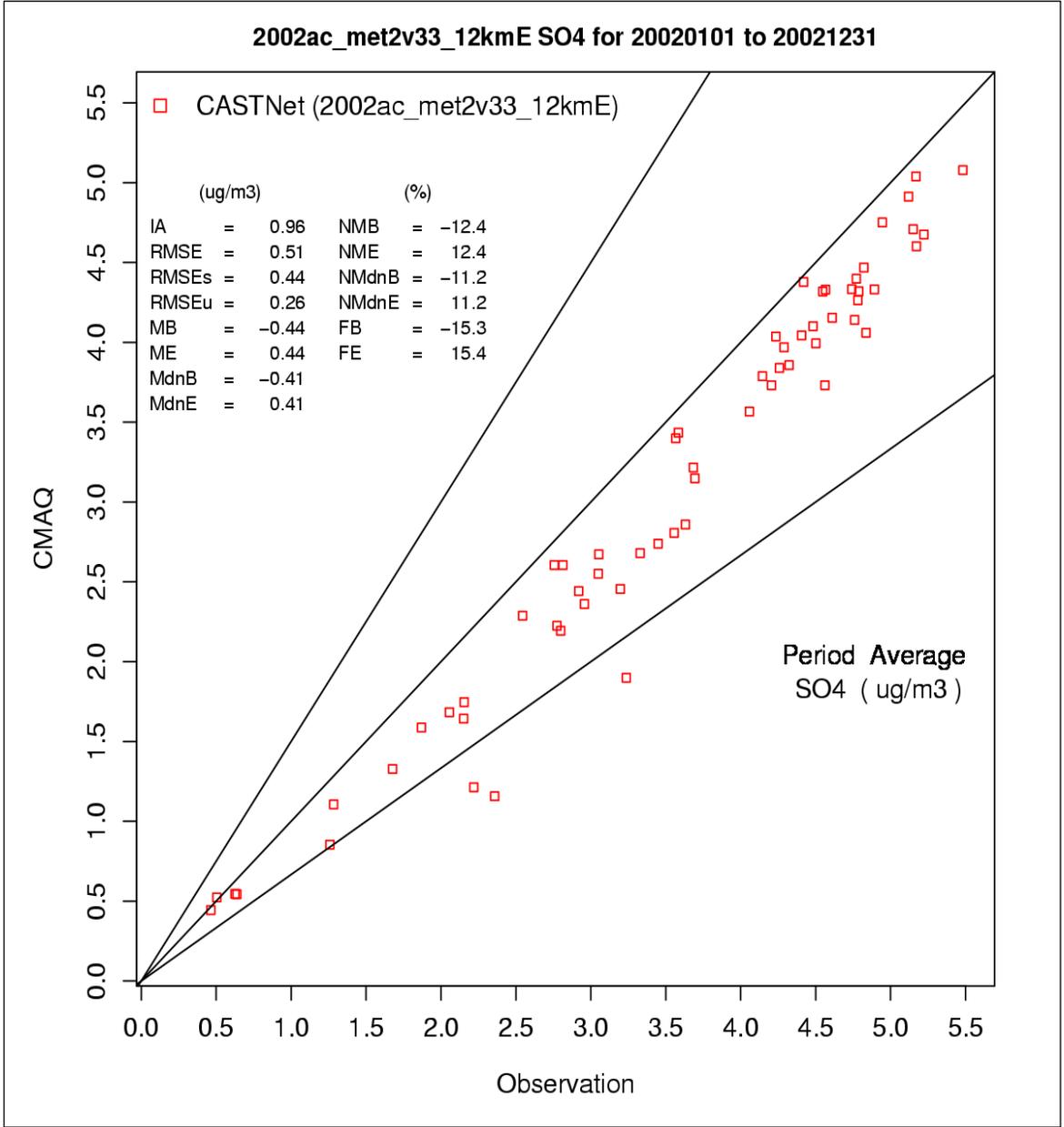
14 The AAPI relies on CMAQ for the sulfur and nitrogen transference ratios and  $\text{NH}_x$   
15 deposition. The model evaluation results, including the ammonia and wet precipitation  
16 treatments, reflect a continual process of model improvement designed to ingest the latest  
17 science within a framework links a myriad of atmospheric and surface processes across multiple  
18 pollutant species. While this document focuses in on the more direct processes affecting N and  
19 S deposition, these modifications are incorporated with the philosophy that the best science is  
20 being adopted and they in turn support the overall improvement of the models' treatment of all  
21 processes. The inclusion of better chemistry and physics of a particular process acts as an  
22 internal diagnostic tool for other processes that are linked throughout the model framework  
23 through basic conservation of mass principles. With respect to the AAPI, the CMAQ model  
24 must be relied on to provide the spatial flexibility attendant with a national standard. As the  
25 model continually adopts the best science, confidence in relevant CMAQ generated AAPI  
26 parameters is raised for both near term and future scenarios.

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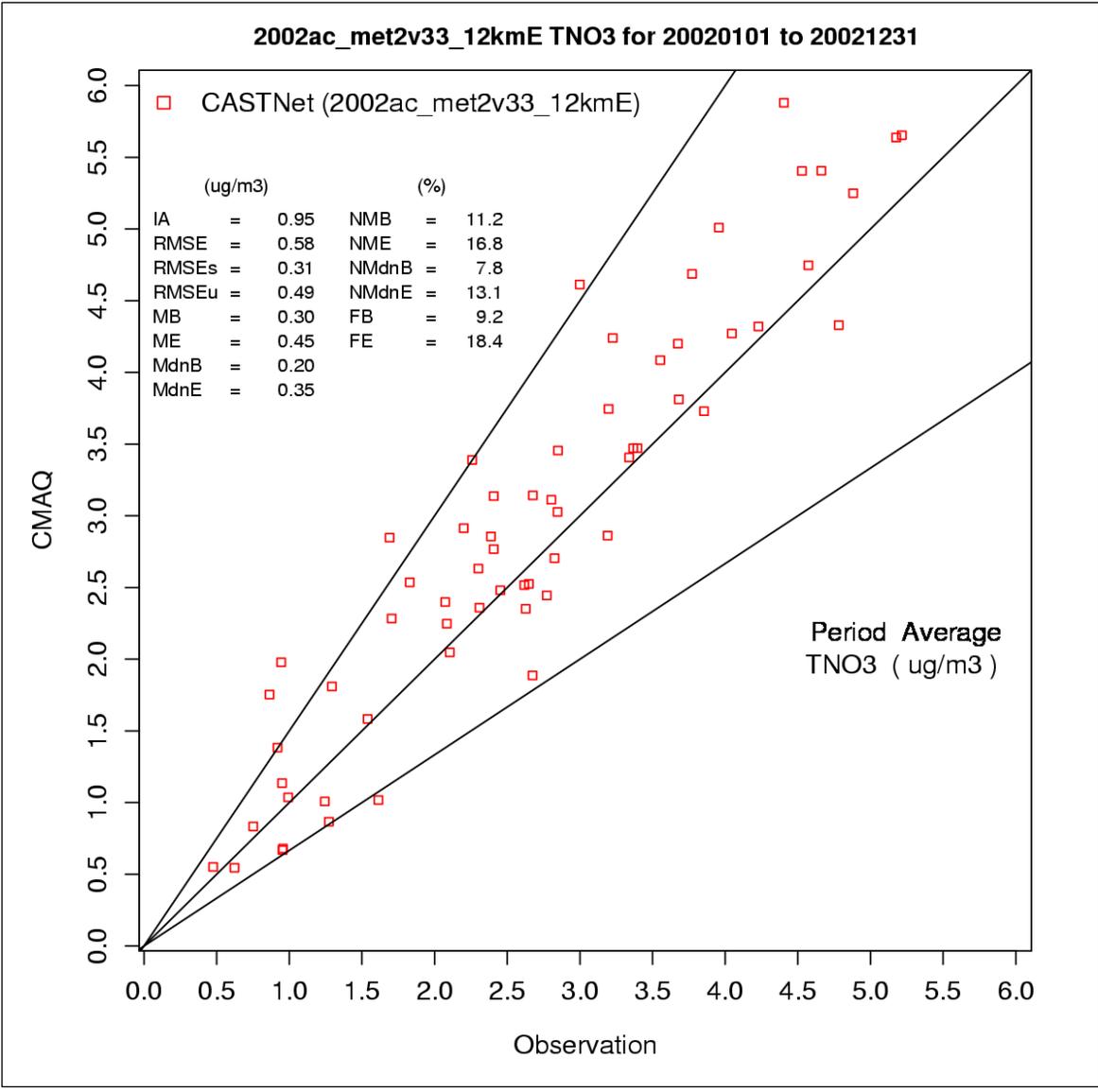
20 **Figure 7-1 2002 CMAQv4.6 annual average SO<sub>2</sub> predicted concentrations versus**  
 21 **observations at CASTNet sites in the eastern domain (note, units are in**  
 22 **actual mass for SO<sub>2</sub>, including oxygen).**

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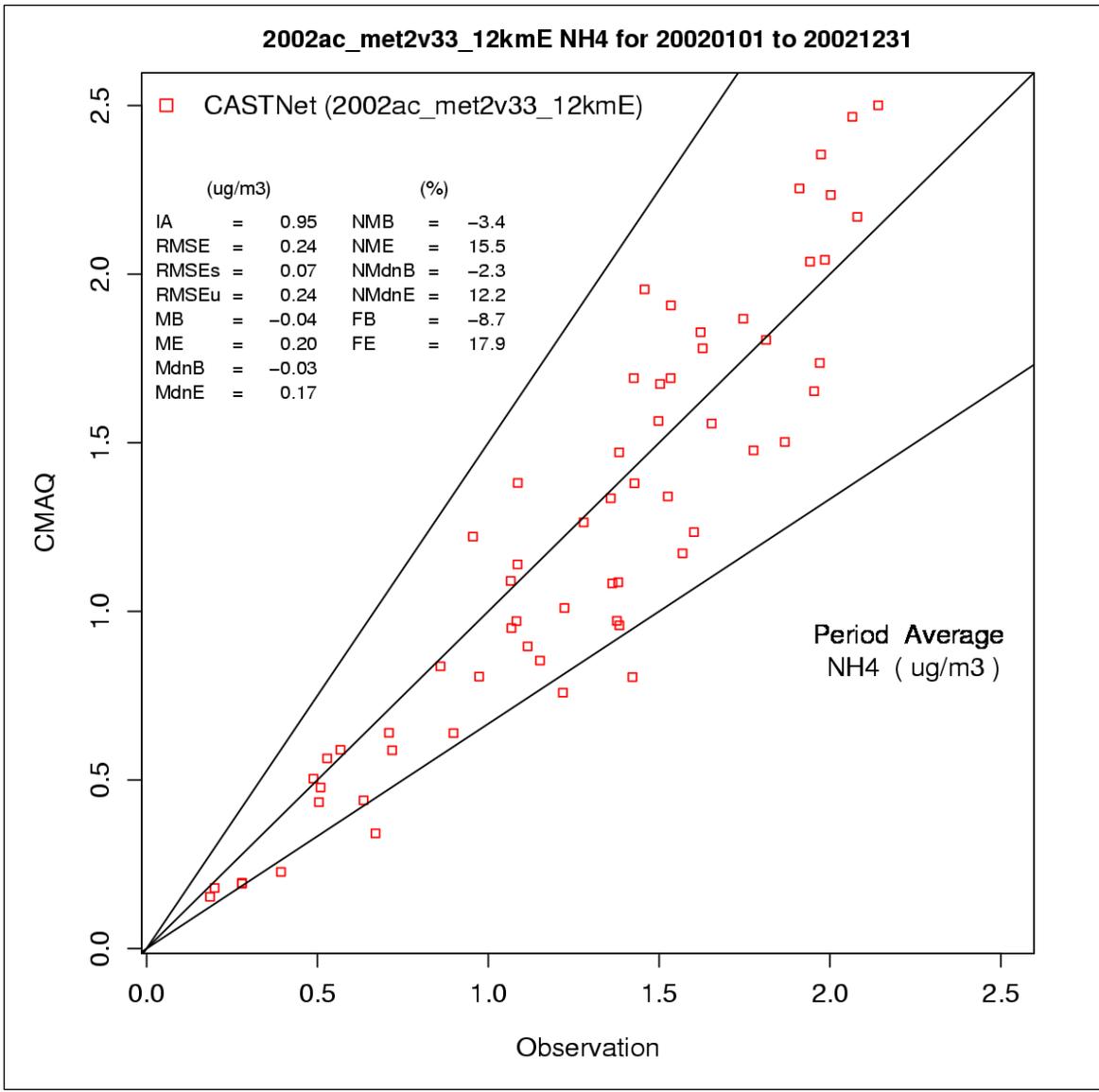
22 **Figure 7-2 2002 CMAQv4.6 annual average SO<sub>4</sub><sup>2-</sup> predicted concentrations versus**  
 23 **observations at CASTNet sites in the eastern domain (note, units are in**  
 24 **actual mass for SO<sub>4</sub>, including oxygen).**

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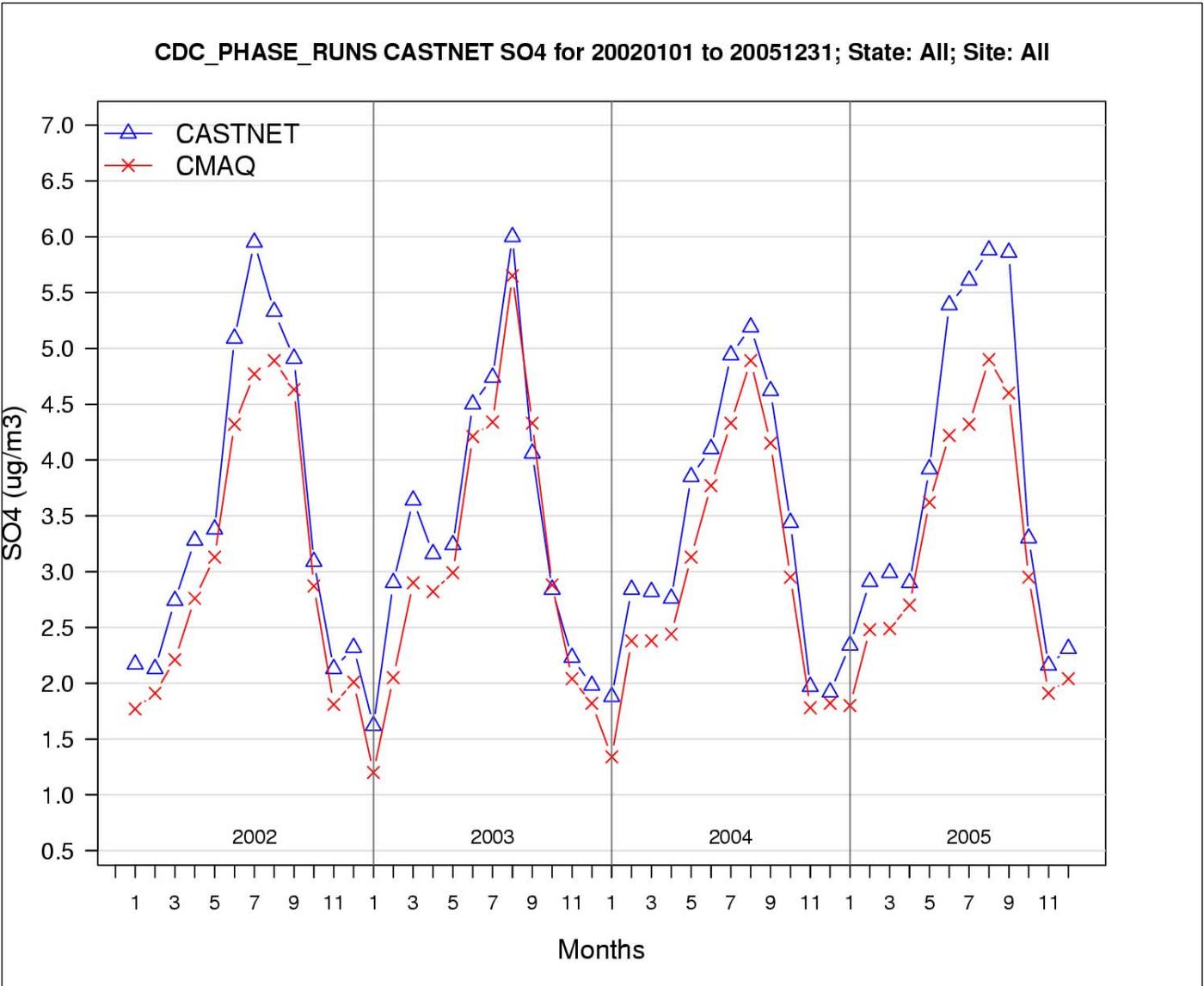
20 **Figure 7-3 2002 CMAQv4.6 annual average TNO<sub>3</sub> predicted concentrations versus**  
 21 **observations at CASTNet sites in the eastern domain (note, units are in**  
 22 **actual mass for NO<sub>3</sub>, including oxygen).**

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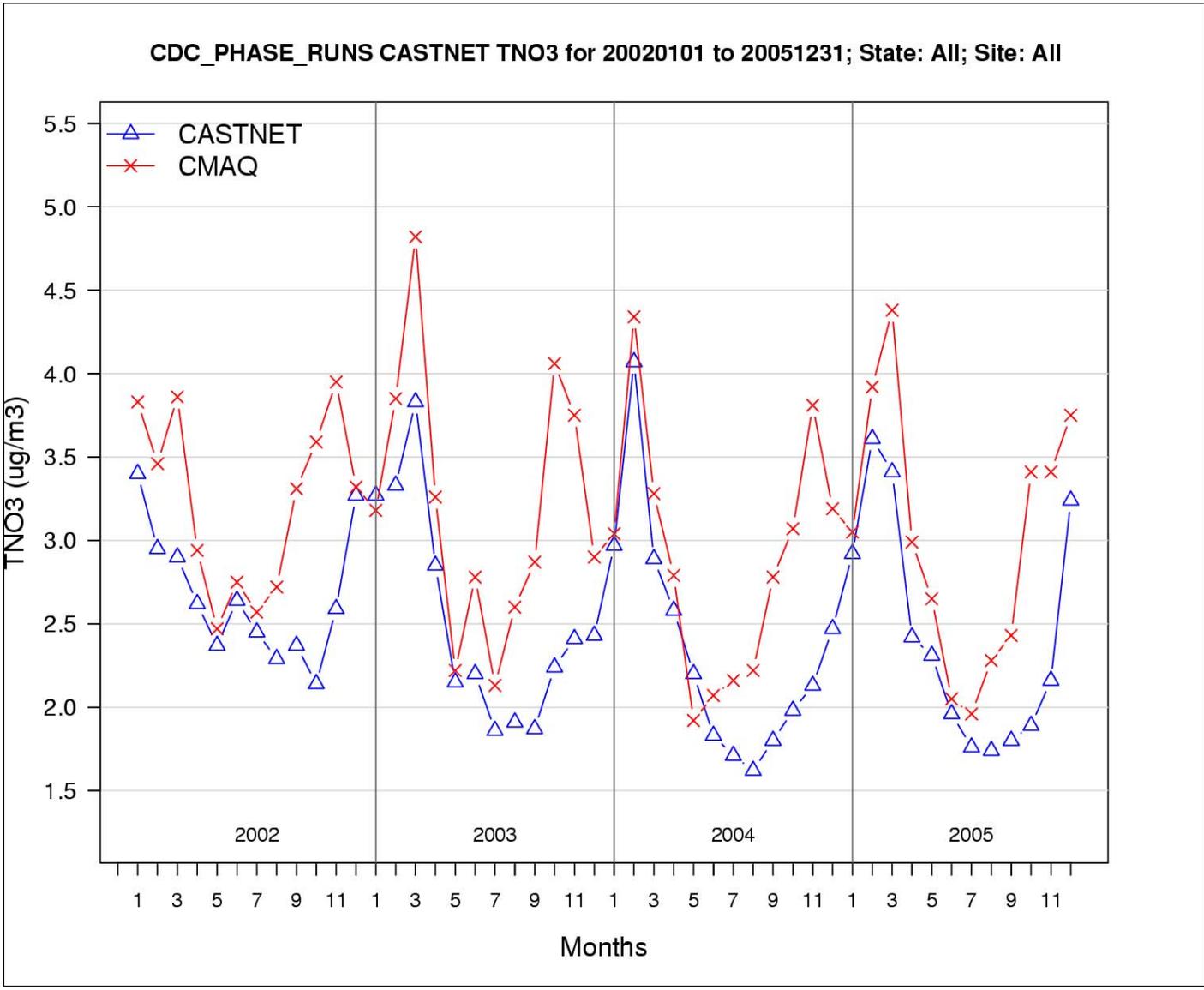
22 **Figure 7-4. 2002 CMAQv4.6 annual average NH<sub>4</sub><sup>+</sup> predicted concentrations versus**  
 23 **observations at CASTNet sites in the eastern domain (note, units are in**  
 24 **actual mass for NH<sub>4</sub>, including hydrogen).**

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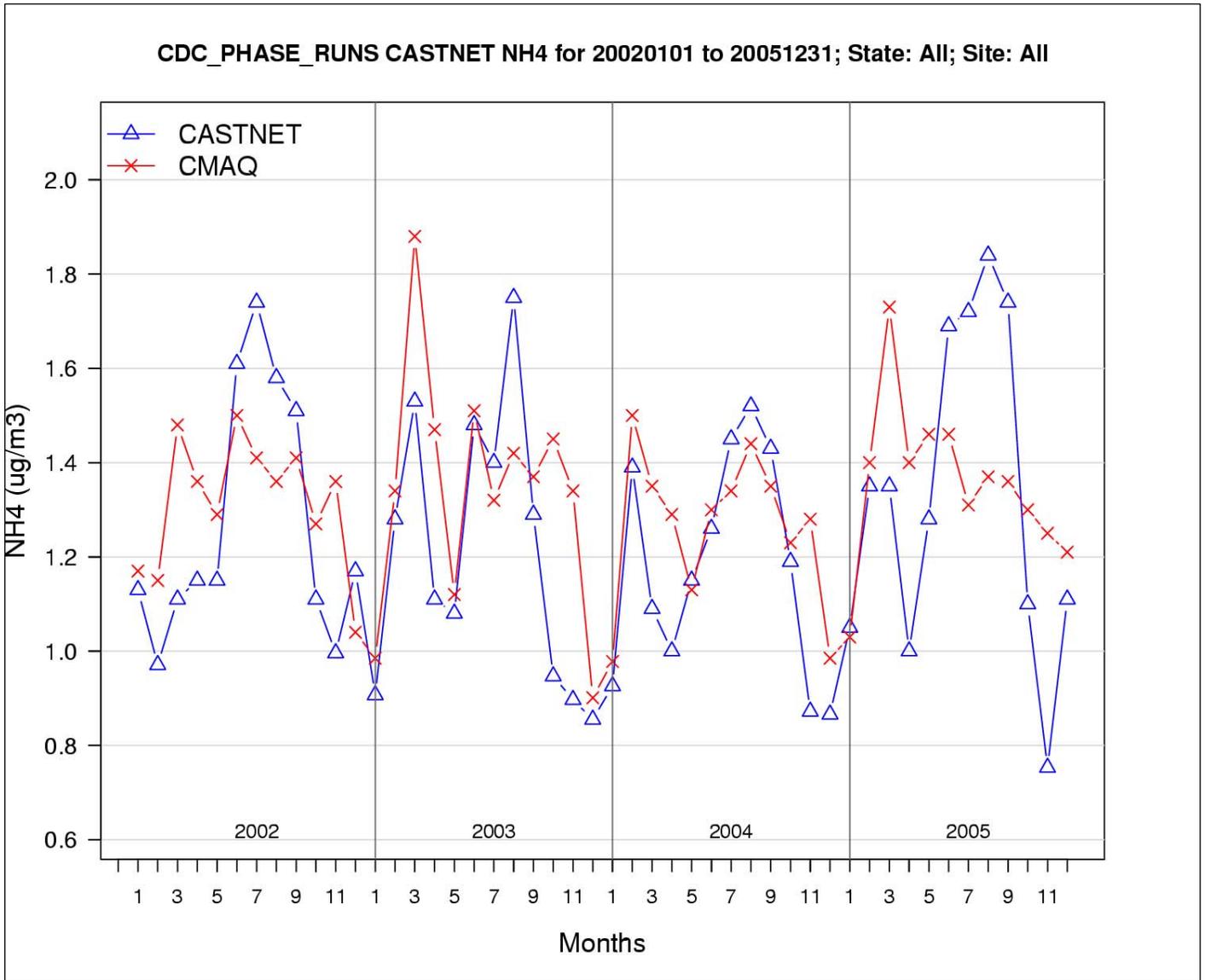
22 **Figure 7-5. 2002–2005 Domain-wide average SO<sub>4</sub><sup>2-</sup> predicted concentrations and**  
23 **observations by month at CASTNet Sites in the eastern domain (note, units**  
24 **are in actual mass for SO<sub>4</sub>, including oxygen) .**

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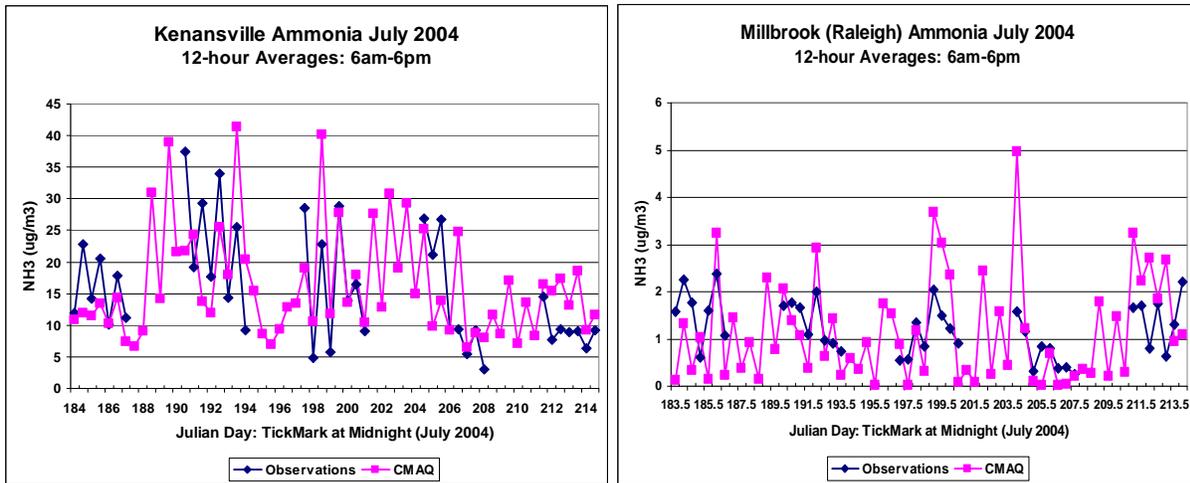
21 **Figure 7-6. 2002–2005 Domain-wide average TNO<sub>3</sub> predicted concentrations and**  
22 **observations by month at CASTNet sites in the eastern domain (note, units**  
23 **are in actual mass for NO<sub>3</sub>, including oxygen).**

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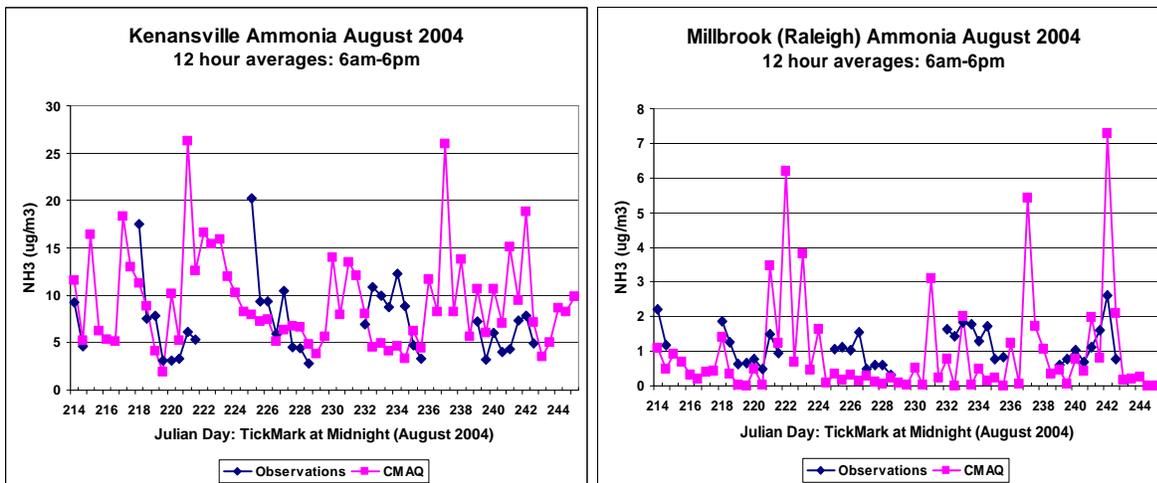


21 **Figure 7-7** 2002–2005 Domain-wide average  $\text{NH}_4^+$  predicted concentrations and  
22 observations by month at CASTNet sites in the eastern domain (note, units  
23 are in actual mass for  $\text{NH}_4$ , including hydrogen).

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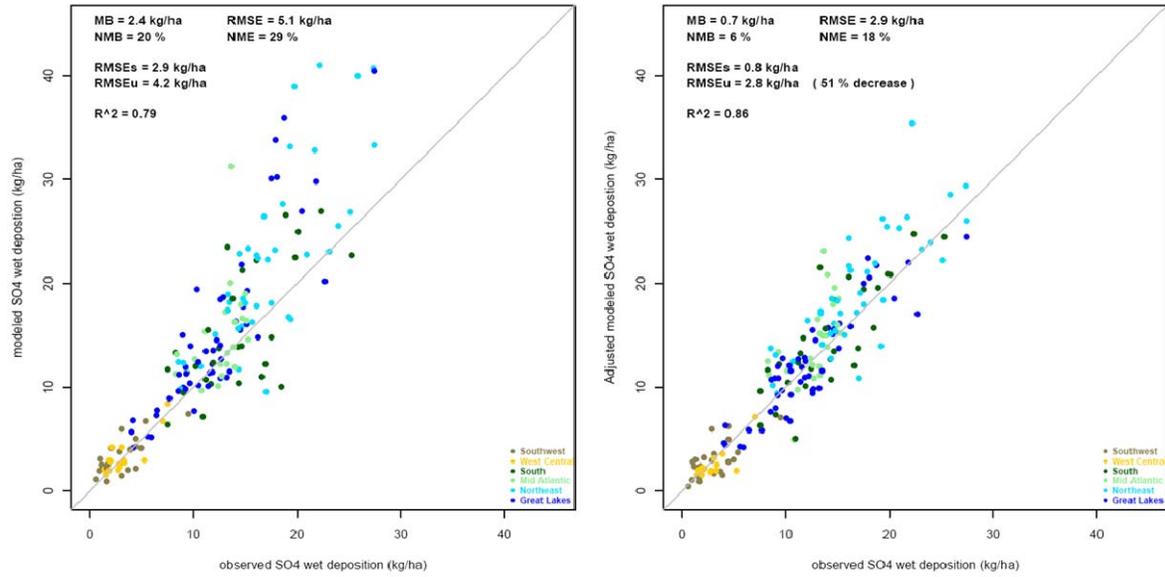


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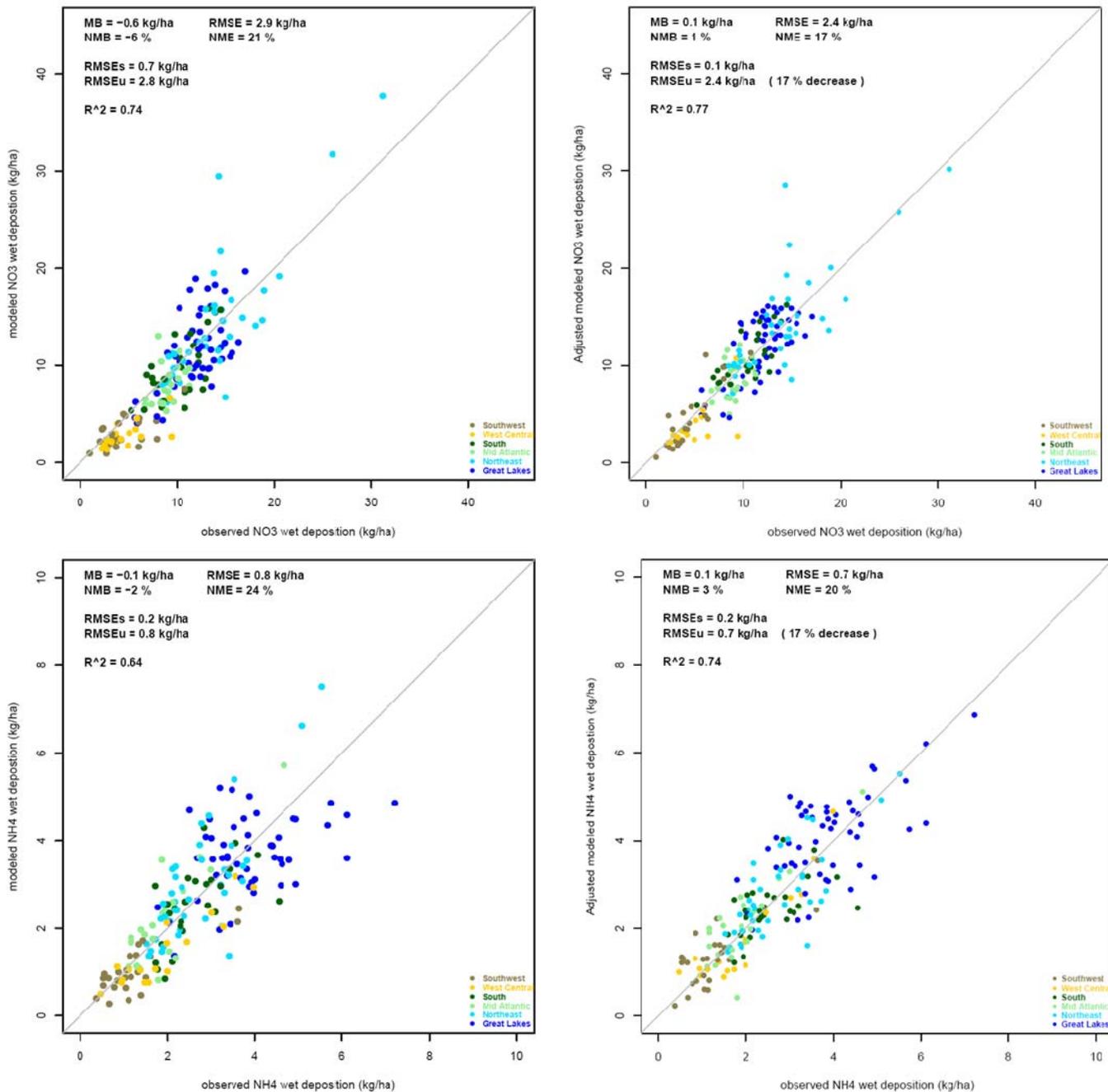
6 **Figure 7-8. Comparison of CMAQ predictions and measurements for 12-hour (6am-**  
7 **6pm) average NH<sub>3</sub> concentrations, with a monitoring cycle of 4 days on and**  
8 **4days off, at a high emission site (Kenansville) and a low emission urban site**  
9 **(Raleigh) in North Carolina compared to CMAQ for July 2004 (top) and**  
10 **August 2004 (bottom), from Dennis et al., 2010 (note, units are in actual mass**  
11 **for NH<sub>3</sub>, including hydrogen).**



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**Figure 7-9 Unadjusted (left) and PRISM (right) adjusted CMAQ annual wet deposited sulfate for 2002 (note, units are in actual mass for SO<sub>4</sub>, including oxygen).**

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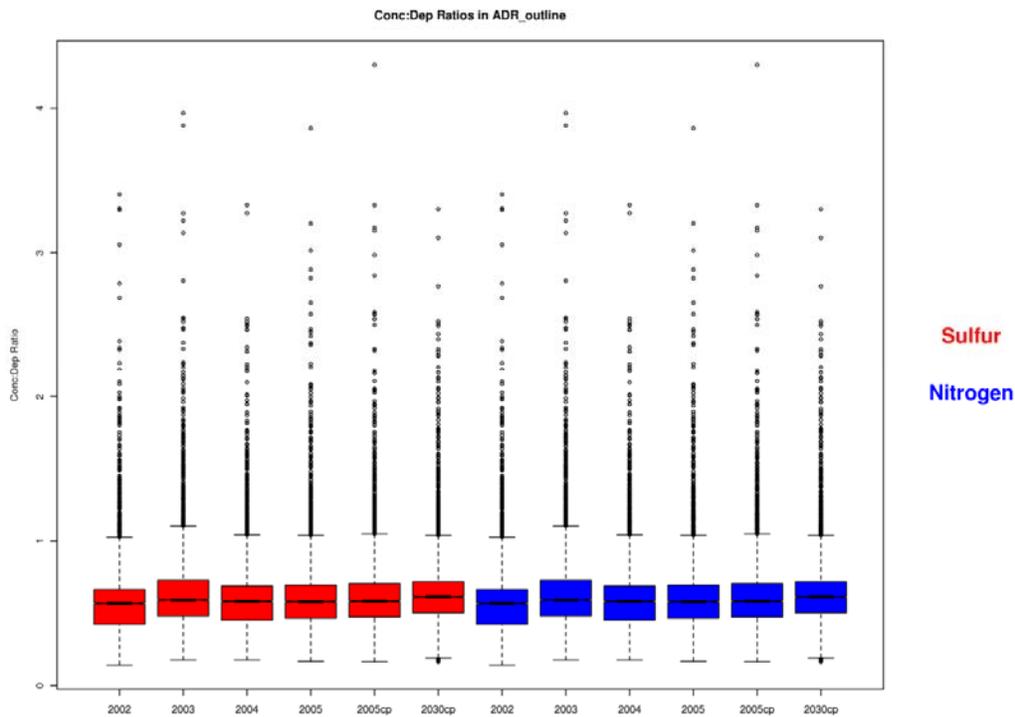
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3 **Figure 7-10 Unadjusted (left) and PRISM and bias (right) adjusted CMAQ annual wet**  
4 **deposition of nitrate (top) and ammonium (bottom) (note, units are in actual**  
5 **mass for NH<sub>4</sub> and NH<sub>3</sub>, including hydrogen).**

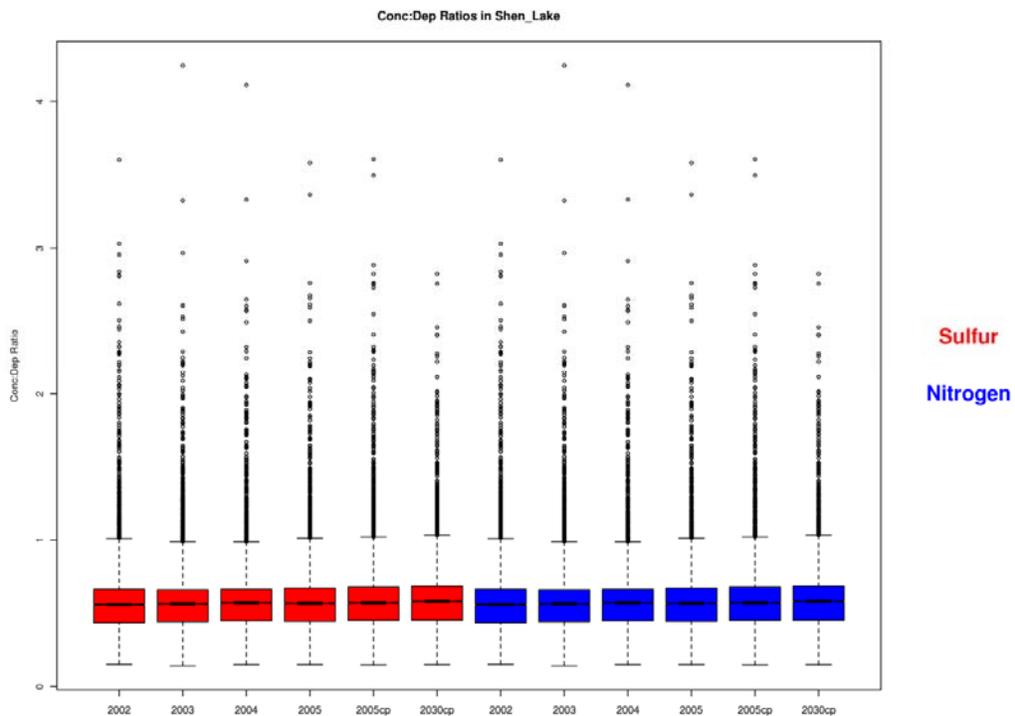
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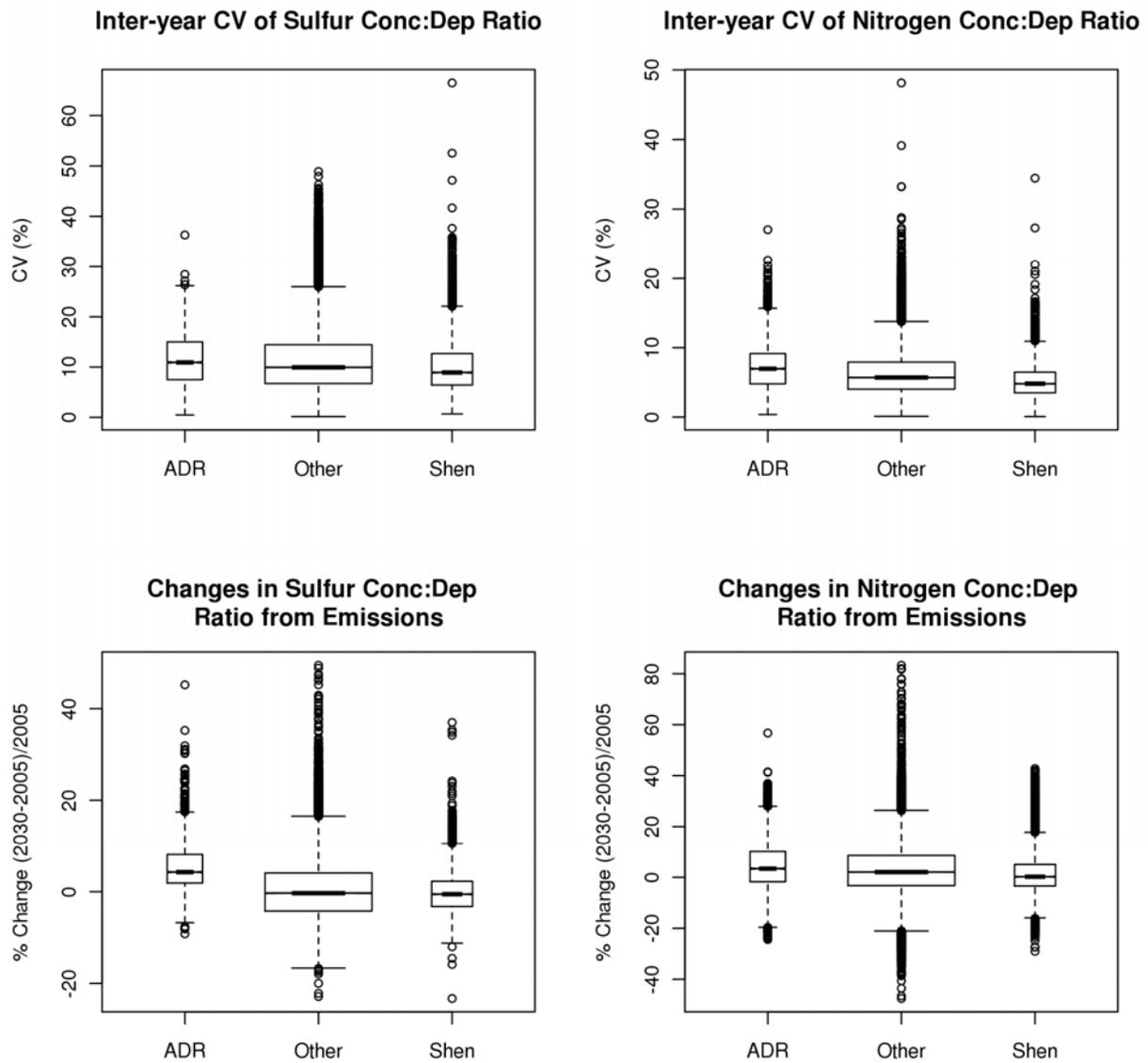


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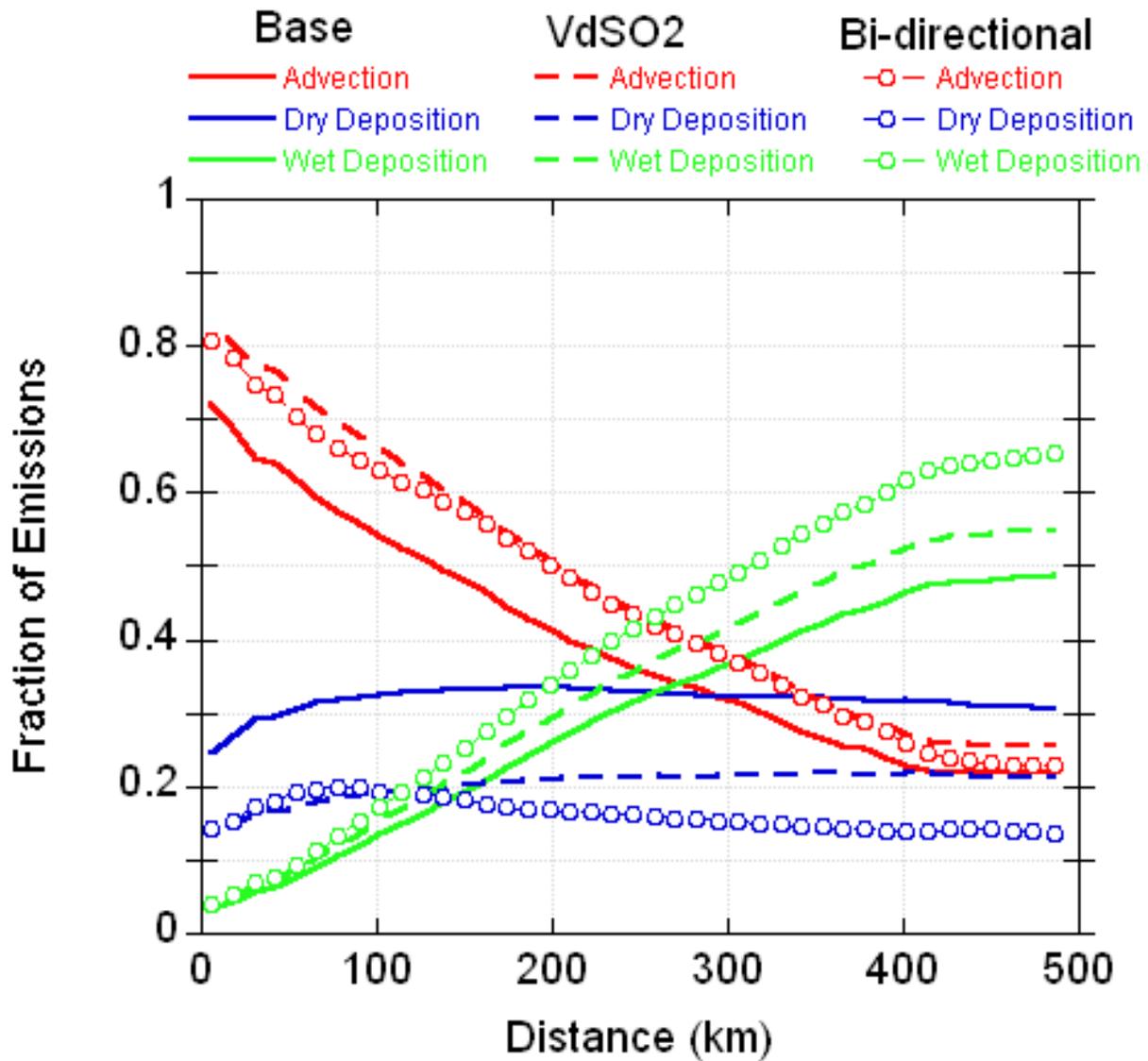
4 **Figure 7-11 Spatial and interannual variability of inverse deposition transferance ratios,**  
5  **$1/T_{SOx}$  and  $1/T_{NOy}$ , for Adirondack (top) and Shenandoah case study areas.**



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**Figure 7-12. Summary of inter-annual and emissions sensitivity variability of sulfur and nitrogen deposition transference ratios.**

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3 **Figure 7-13. Cumulative regional NH3 budget of advection, wet- and dry deposition,**  
4 **calculated for an expanding box starting at the high-emitting Sampson**  
5 **County NC cell (from Dennis et al, 2010)**

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1 **7.5. Sensitivity of AAPI to component parameters**

2 An elasticity analysis was applied to investigate sensitivity of the AAPI to its components  
3 (Appendix A). The means, medians and quartiles of the AAPI component variables were based  
4 on the range variable values across ecoregions that overlapped with the CMAQ domains.  
5 Elasticities measure the percent change in the AAPI for a 1% change in the AAPI parameters:  $Q$ ,  
6  $N_{eco}$ ,  $NH_x$ ,  $BC_0^*$ ,  $T_{NOy}$ ,  $T_{SOx}$ ,  $NO_y$ , and  $(SO_2 + SO_4)$ .

7 
$$AAPI = \frac{1}{Q} N_{eco} + BC_0^* - \frac{1}{Q} NH_x - \frac{1}{Q} [T_{NOy} \cdot NO_y + T_{SOx} \cdot (SO_4 + SO_2)]$$

8 The elasticity results identified significance for all the AAPI parameters. Detailed  
9 results are provided in Appendix A. Base cation weathering,  $BC_0^*$ , and hydraulic flow rate,  $Q$ ,  
10 exerted strong influence on AAPI, an expected result given the explicit dependency evident in  
11 the AAPI expression. The transference ratios for  $NO_y$  ( $T_{NOy}$ ) and  $SO_x$  ( $T_{SOx}$ ) exhibited  
12 relatively less influence on AAPI calculations than all other parameters when evaluated at means  
13 of the variables. However, in some locations, when evaluated at other values of the variables,  
14 AAPI can be more sensitive to the deposition transformation ratios.

15 These results suggest focusing on the uncertainties in the non-atmospheric inputs,  
16 including base cation weathering and runoff rates, and the implications of those uncertainties in  
17 setting an AAPI that will have a high likelihood of providing the targeted level of protection.  
18 An analysis of variance (ANOVA) analysis of the AAPI parameters will be added to the final  
19 PA.

20

21 **7.6 Uncertainty in Critical Load and ANC modeling**

22 **7.6.1 MAGIC modeling**

23 An extensive uncertainty analysis of the MAGIC model was conducted as part of the  
24 REA, and documented in Appendix 4 of the REA. This uncertainty analysis included  
25 comparison of MAGIC outputs with observed water chemistry and ANC values. The  
26 uncertainty analysis also included an approach for generating confidence intervals for predicted  
27 ANC, using ensembles of model results based on alternative model calibration methods.

1           The model performance comparisons documented in Appendix 4 of the REA show close  
2 correspondence between simulated and observed annual average surface water SO<sub>4</sub>, NO<sub>3</sub>, and  
3 ANC during the model calibration period for 44 lakes in the Adirondacks Case Study Area and  
4 60 streams in the Shenandoah Case Study Area. These comparisons are reproduced in Figures 7-  
5 14 and 7-15. Comparisons in the ability of MAGIC to reproduce the temporal pattern of ANC  
6 for individual lakes was also assessed, and the model does reasonably well at matching the  
7 pattern of ANC, although the fit is not as good as during the model calibration period.

8           The estimated confidence bounds on predicted ANC suggest that the 95 percent upper  
9 confidence bound is on average 10 percent higher in lakes, and 5 percent higher in streams. This  
10 suggests relatively low uncertainty introduced by the MAGIC modeling assumptions. MAGIC  
11 modeling is used in developing the estimates of base cation weathering for comparison to the F-  
12 factor approach described in Chapter 5.

### 13 **7.6.2 SSWC modeling**

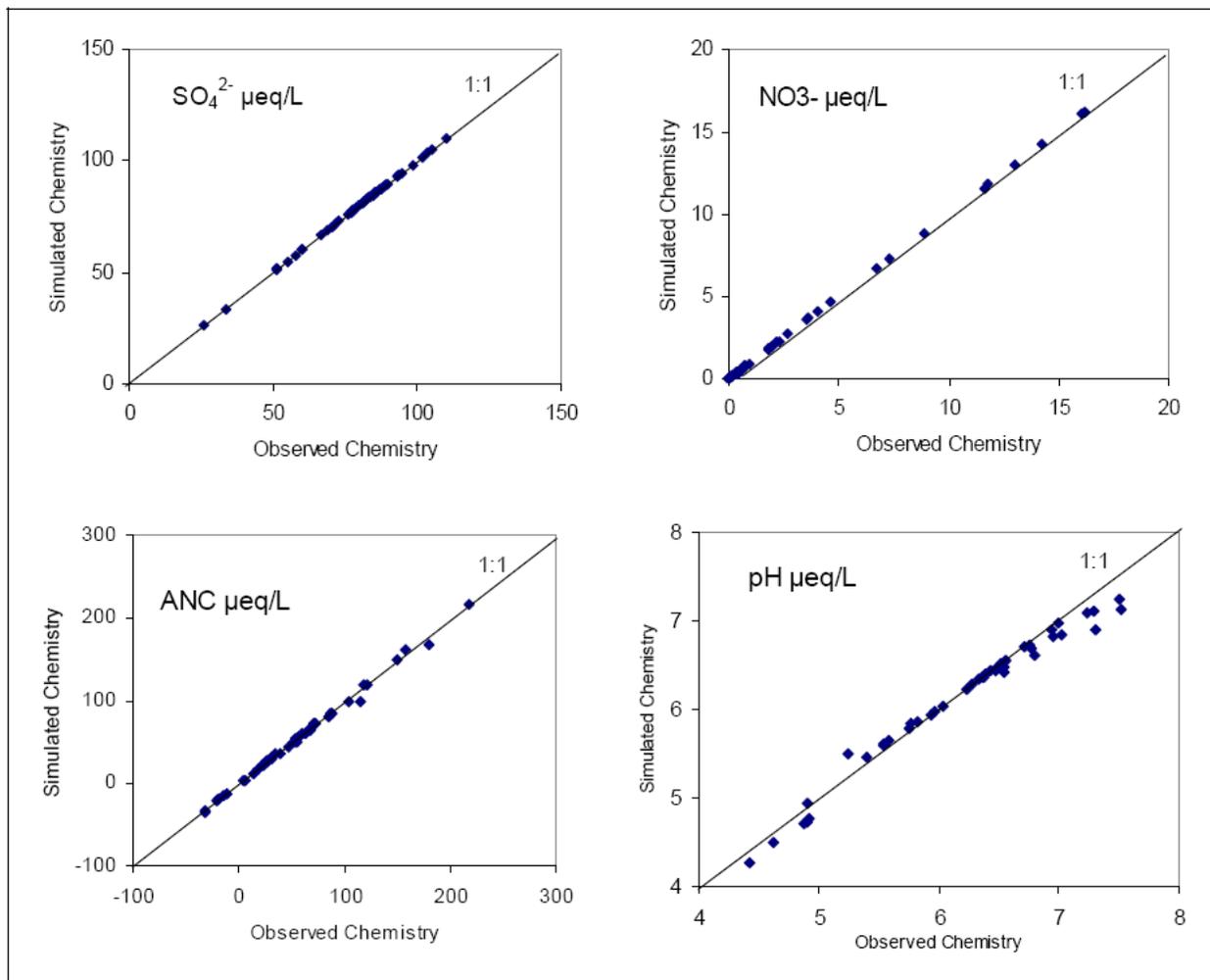
14           As stated in Appendix 4 of the REA, uncertainties in some elements of the SSWC  
15 modeling are not well understood. The version of the SSWC model used here uses the F-factor  
16 approach to estimate the preindustrial base cation supply for a given catchment. While this  
17 approach has been widely applied in Canada and Europe, it has only been used in a few cases  
18 within the United States and its assumptions and parameters have not been fully evaluated for  
19 aquatic systems. The natural or preindustrial catchment supply of base cations (i.e. weathering  
20 rates) has the most influence on the critical load calculation and also has the largest uncertainty  
21 (Li and McNulty, 2007). The uncertainty and ability to accurately estimate this parameter has  
22 not fully been evaluated and its uncertainty is unknown. It is important to note that for the  
23 United States, there is only one study for surface waters critical loads that compared steady-state  
24 and dynamic models and different steady-state approaches (MAGIC and F-factor) (Holdren et al.  
25 1992) other than what is presented in Chapter 5. Holdren et al. 1992 compared critical loads  
26 calculated by the steady-state MAGIC and the SSWC F-factor model for lakes in the Northeast.  
27 In this study, steady-state MAGIC model yielded critical load values that show the same general  
28 trend and on average were 14 kg/(ha-yr) SO<sub>4</sub> higher than those from the SSWC F-factor  
29 approach, which is consistent with results, presented in Chapter 5. The two models converge at

1 low critical, but diverge as the buffering potential for watersheds increase, as indicated by  
2 increasing critical loads.

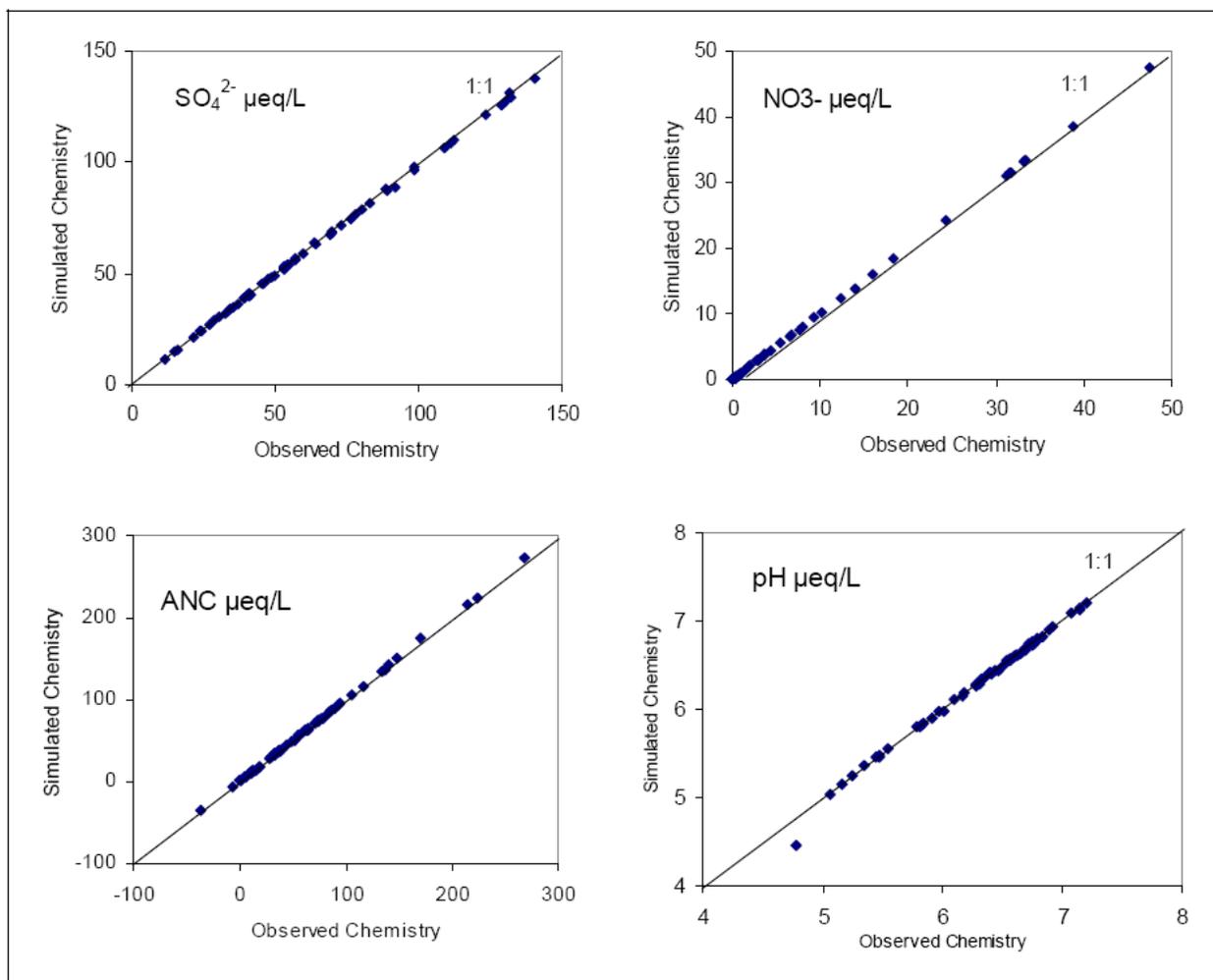
3 The REA conducted an uncertainty assessment using Monte Carlo simulation methods to  
4 characterize the uncertainty in estimated critical loads using the SSWC, varying a number of  
5 important inputs including runoff rates, water chemistry variables, and acid deposition. The  
6 coefficients of variation (CV) for the estimated critical loads (standard deviation divided by the  
7 mean) were calculated for each lake in the study as a measure of relative uncertainty.

8 The results of this uncertainty analysis show that the coefficients of variation are on  
9 average very low for target ANC values within the range we are recommending (20 to 50  $\mu\text{eq/L}$ ).  
10 The CVs for critical loads are only 5% and 9% for critical load limits of 20 and 50  $\mu\text{eq/L}$ ,  
11 respectively. Although the average CV is relatively small for the population of sites modeled,  
12 individual site CV can vary from 1% to 45%. This difference is due to the high degree of  
13 uncertainty in site specific parameters for particular sites.

14 These analyses suggest that uncertainties introduced in the AAPI directly by the SSWC  
15 Factor model are likely to be moderate. Additional uncertainties are introduced by the  
16 generalization of the F-factor approach to estimate critical loads in locations where F-factors  
17 have not been developed.



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 2 **Figure 7-14 Simulated versus observed annual average surface water SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup>, ANC,**  
 3 **and pH during the model calibration period for each of the 44 lakes in the**  
 4 **Adirondacks Case Study Area. The black line is the 1:1 line. (Source:**  
 5 **reproduced from REA, Appendix 4, Figure 1.1-1)**



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 2 **Figure 7-15. Simulated versus observed annual average surface water SO<sub>4</sub><sup>2-</sup>, NO<sub>3</sub><sup>-</sup>, ANC,**  
 3 **and pH during the model calibration period for each of the 60 streams in the**  
 4 **Shenandoah Case Study Area. The black line is the 1:1 line. (Source:**  
 5 **reproduced from REA, Appendix 4, Figure 1.1-2)**

6 **7.7. Modeling and Data Gaps (To be expanded in final PA)**

7 **Atmospheric and deposition processes.** The previous section introduced two important  
 8 enhancements regarding the treatment of wet precipitation and the bi-directional flux of  
 9 ammonia. The interest in deposition of sulfur and nitrogen raises the potential importance of  
 10 occult (cloud and fog related processes) deposition associated with mists and clouds, which may  
 11 be particularly relevant for aquatic acidification of high elevation watersheds. Occult deposition  
 12 currently is in the early stages of development within the CMAQ framework.

1 Lightning generated NO<sub>x</sub> emissions have been an active area of research over the last  
2 decade and approaches that incorporate lightning count data and estimated NO<sub>x</sub> generation based  
3 on satellite measurements and aircraft campaigns have been tested in modern air quality models,  
4 including CMAQ. Lightning NO<sub>x</sub> is hypothesized to increase upper tropospheric ozone levels  
5 and wet nitrogen precipitation, with relatively negligible impact on near surface ambient nitrogen  
6 patterns. It is anticipated that CMAQ will incorporate lightning NO<sub>x</sub> for EPA assessments in the  
7 2012 timeframe.

8 Interest in organic bound nitrogen has increased based on NADP measurements  
9 suggesting that organic nitrogen contributes as much as 30% of the total nitrogen in precipitation  
10 samples. Significant uncertainties regarding the origin and composition of organic nitrogen  
11 (Altieri et al., 2009) suggest a need for research to improve our understanding of organic  
12 nitrogen prior to developing parameterizations in air quality models. Questions regarding the  
13 the relative contribution of anthropogenic or natural sources as well as the effects of re-  
14 entrainment from the surface require attention.

15 **Atmospheric Observations.** Chapter 4 addresses the current state of atmospheric  
16 observations relative to the NO<sub>x</sub>/SO<sub>x</sub> secondary standard and Chapter 8 addresses preliminary  
17 recommendations for monitoring methods. This new standard poses measurement resource  
18 challenges as the current networks, with the exception of CASTNET and some National Park  
19 Service (NPS) efforts, are deficient in spatial coverage relevant to anticipated acid sensitive areas  
20 and the specific measurement needs related to NO<sub>y</sub>, speciated NO<sub>y</sub> and ammonia and  
21 ammonium.

22 **Source emissions.** Anthropogenic emissions of nitrogen oxides (NO and NO<sub>2</sub>) and  
23 sulfur dioxide generally are believed to be well characterized as the major contributors of NO<sub>x</sub>  
24 and SO<sub>2</sub> from energy generation and transportation sectors have a history of continuous  
25 improvements of emissions modeling as well as direct emission measurements for major power  
26 generating units. Greater uncertainty resides in natural emissions of NO<sub>x</sub> from lightning  
27 processes (discussed above) and soil and agricultural related phenomena. Both NO<sub>x</sub> and  
28 ammonia emissions are subject to re-emission after deposition as part of the complex cycling of  
29 nitrogen in soils and biota. Characterizing the variety of agricultural practices that impact both  
30 ammonia and NO<sub>x</sub> is complicated by the dispersed nature of agriculture processes as well as the

1 influence of various meteorological factors on relevant biogeochemical processes controlling  
2 transformation and removal of nitrogen species.

3 **Ecosystem processes and surface water observations.** [To be completed in final PA]

4 The critical load modeling approaches that produced the N/S deposition tradeoff curves require a  
5 variety of input data depending on the approach chosen. In general terms, the availability of  
6 watershed related deposition, soil and vegetation characteristics and surface water chemistry  
7 determine the approach taken. There is a relatively extensive source of data for critical load  
8 modeling in the Eastern U.S., as illustrated by the frequent reliance on the Adirondack and  
9 Shenandoah Case studies. For this PA, critical load estimates were developed for national level  
10 coverage, largely through SSWC modeling relying on surface water data. Several ecoregions  
11 included an extremely small sample size of critical load estimates that challenged the  
12 development of a national scale assessment of acid sensitive areas. A more thorough  
13 characterization of nitrogen retention, dissolved organic carbon, soil chemistry in all acid  
14 sensitive areas would lead to reduced uncertainties in applying the AAPI as well as future  
15 considerations for standards that incorporate terrestrial acidification and nutrient enrichment  
16 effects.

17 **7.8. Summary and Conclusions**

18 Uncertainty and natural variability exist in all of the components of the structure of the  
19 NO<sub>x</sub> and SO<sub>x</sub> standard introduced in this PA, and should be considered in establishing the level  
20 of the AAPI. A summary of the relative uncertainties of these components is provided in table  
21 7-1 (To be added). On balance, the confidence level in the information and processes associated  
22 with the linkages from ecological effects to atmospheric conditions through deposition and  
23 ecosystem modeling is very high. The considerable body of evidence is conclusive with regard  
24 to causality between aquatic acidification and biological and ecological effects. Confidence in  
25 the linkage associating aquatic acidification and ANC is extremely high, as the aquatic chemistry  
26 describing this relationship, while nonlinear, is relatively simple with regard to chemical species  
27 and reactions. The relationships between deposition and ANC, while complicated by a variety  
28 of biogeochemical and hydrological processes and data requirements within watersheds, are well  
29 established and the critical load models have been thoroughly vetted through the scientific  
30 community with a demonstrated level of successful evaluation. The linkages between ambient

1 concentrations of relevant species and deposition is best handled through air quality modeling  
2 systems like CMAQ. The relationship between concentrations and deposition loads is well  
3 characterized by these models, which are constrained by mass balance principles. While much  
4 of the physical and chemical processing that determine concentrations and consequent deposition  
5 is interwoven with numerous fundamental processes characterizing mass transport and  
6 atmospheric chemical oxidation, the science is relatively mature with years of applications and  
7 continued evolution of the models. The specific processes guiding nitrogen and sulfur  
8 chemistry and deposition are relatively simple. More challenging is the ability to parameterize  
9 processes at the air-surface interface which guide the estimation of deposition velocities and the  
10 re-emission of certain species, as well as many of the area wide natural processes and  
11 agricultural practices which influence emissions of oxidized and reduced forms of nitrogen.

12 The variety of uncertainty, variability and sensitivity analyses included in this chapter  
13 have been conducted under the assumption that the basic model construct is solid, as discussed  
14 immediately above, and are used to inform conclusions regarding the level of the AAPI that  
15 incorporate consideration of uncertainty. These analyses are also useful in guiding  
16 implementation efforts related to future monitoring, emissions and model process improvements.

17 The influence of uncertainty on the level of the AAPI can be thought of as reducing or  
18 increasing relative stringency of the level to increase the likelihood that requisite protection of  
19 public welfare is provided. Throughout these discussions there is no apparent directional bias  
20 in the uncertainty regarding the biological, chemical and physical processes incorporated in the  
21 AAPI. From the perspective of valuation of ecosystem services, the estimates generally are  
22 believed to be biased low, meaning the values of reaching a target level of protection are  
23 underestimated. However, quantification of these values is perhaps the most uncertain of all  
24 aspects considered. Consequently, the level of the AAPI should be relatively high in a buffering  
25 context to account for the existence of uncertainties in several components. In addition to, but  
26 related to these uncertainties discussions, are considerations of time lag to reach a target level  
27 ANC due to ecosystem response dynamics, as well the uncertainties in the severity and  
28 prevalence of episodic events. Both of these considerations suggest support for an AAPI that is  
29 somewhat higher than the target ANC supported by the specific evidence and risk information.

30

1 **Table 7-2. Summary (Incomplete) of Qualitative Uncertainty Analysis of Key Modeling Elements in the NO<sub>x</sub>/SO<sub>x</sub> AAPI.**

Source	Description	Potential influence of uncertainty on risk estimates		Knowledge-Base uncertainty*	Comments (KB: knowledge base, INF: influence of uncertainty on AAPI estimates)
		Direction	Magnitude		
<b>Major elements (and sub-models) of the ecological effects to ambient concentration framework</b>					
Biological/ecosystem response to acidification	Clear associations between aquatic acidification (pH, elevated Al) and adverse ecosystem effects (fish mortality, decreased species diversity)	Both	Low	Low	
Linkage between direct acidification species to ecological indicator (ANC)	The relationships across ANC, pH and dissolved Al are controlled by well defined aquatic equilibrium chemistry	Both	Low	Low	ANC is the preferred ecosystem indicator as it has a direct relationship with pH and the deposition species relevant to the NO <sub>x</sub> /SO <sub>x</sub> standard.
Linkage between ecological indicator and adverse ecological effects	Direct associations between ANC and fish mortality and species diversity	Both	Low-medium	Low	Although the pH dependency on ANC is nonlinear, it is always directionally consistent. In extremely low and high ANC environments the relationship is of minimal value as catchments are in relatively "less sensitive" regimes due to natural conditions or extreme anthropogenic influence (i.e., acid mine drainage). In sensitive areas of concern the relationship essentially is similar to the relationships between direct acidification species and adverse effects.
Deposition to ANC linkage through Critical Load modeling	Both MAGIC and Steady State critical load models are applied to determine critical load models. The Steady State critical load model formulation is used as the foundation for deriving the	Both	Low	Low	The model formulation is well conceived and based on a substantial amount of research and applications available in the peer reviewed literature. There is greater uncertainty associated with the availability of data to support certain model components.

Source	Description	Potential influence of uncertainty on risk estimates		Knowledge-Base uncertainty*	Comments (KB: knowledge base, INF: influence of uncertainty on AAPI estimates)
		Direction	Magnitude		
	AAPI equation.				
Atmospheric concentrations to Deposition	Deposition is a direct function of ambient concentration, influenced by several processes, and handled in this PA through air quality modeling.	Both	Low	Low	The model design is appropriate given the spatial and temporal complexities that influence deposition velocity, as well as the variety of atmospheric species that generally are not measured. Greater uncertainty resides in the information driving (e.g., ammonia emissions) these calculations and availability of observations to evaluate model behavior.
<b>Sub-components and data of individual models</b>					
Deposition Transference Ratios	CMAQ derived ratio of total oxidized deposition to concentration averaged over one year	both	low	unknown	Transference ratios enable the connection between deposition and the policy relevant ambient air indicators, NOy and (SO <sub>2</sub> + SO <sub>4</sub> ). They are strictly a model construct and cannot be evaluated in a traditional model to observation context. The low sensitivity of these ratios to emission changes and inter annual meteorology combined with low spatial variability indicate that these ratios are necessarily stable.
Additional elements to be added (.BCo, NECO, Q, DOC, benefits, ambient obs, surface water obs, emissions, ....)					

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23

## 8 AMBIENT AIR MONITORING

Ambient air measurements of nitrogen and sulfur species support implementation of this proposed NAAQS secondary standard and improve the information basis for subsequent reviews. These uses extend beyond the basic need to measure the proposed NAAQS indicators,  $\text{NO}_Y$ ,  $\text{SO}_2$  and  $\text{SO}_4$ , in approximate priority order:

- Direct NAAQS comparisons.
- Reduced nitrogen,  $\text{NH}_3$  and  $\text{NH}_4$ , to evaluate CMAQ and other air quality models ability to characterize  $\text{NH}_X$  deposition, a component of the AAPI expression.
- Model and process improvement - In combination with NAAQS indicators and  $\text{NH}_X$ , additional speciated  $\text{NO}_Y$  components including  $\text{HNO}_3$ , PAN,  $\text{NO}_2$ , and NO to continually assess air quality model behavior and associated deposition processes.
- Subsequent NAAQS reviews of the secondary  $\text{NO}_X/\text{SO}_X$  standard as well as related primary and secondary standards reviews (ozone,  $\text{NO}_2$ ,  $\text{SO}_2$  and PM).
- Accountability – Assessing the effectiveness of implemented programs addressing emission strategies to meet attainment of the proposed NAAQS using all noted measurements.

### 8.1 What Are The Appropriate Ambient Air Indicators To Consider In Developing The Standards?

The recommendation of  $\text{NO}_Y$ ,  $\text{SO}_2$  and  $\text{SO}_4$  as the ambient air indicators for the proposed  $\text{NO}_X/\text{SO}_X$  standard was introduced in the first draft of the PAD and endorsed by CASAC (CASAC, 2010). Essentially,  $\text{NO}_Y$ , which is an aggregate of all reactive oxidized nitrogen compounds and the two sulfur species represent the oxidized ambient air species of relevance to the criteria pollutants  $\text{NO}_X$  and  $\text{SO}_X$  with potential to adversely affect acid-base balance in aquatic systems. Contributions of reduced nitrogen, which would not be part of the indicator for the standard under the approach suggested in this PAD, are provided by CMAQ.

1            *Why not use each individual species as indicators?*

2            One could consider using all NO<sub>y</sub> species as NAAQS indicators, requiring, for example,  
3 measurements of the dominant species: HNO<sub>3</sub>, particulate nitrate, true NO<sub>2</sub>, NO, and PAN.  
4 Conceptually, each species would be paired with a species - specific deposition velocity in the  
5 AAPI expression to transfer individual deposition values to ambient values. The attraction of  
6 using individual species would be the reliance on actual deposition velocities that have more  
7 physical meaning in comparison to model constructed transference ratios which aggregate dry  
8 and wet deposition and all nitrogen species. The transference ratio approach does retain the  
9 necessary conservation of mass which underlies virtually all parameterization schemes, but loses  
10 some degree of physical relevance due to use of modeled outputs – an admittedly unique  
11 construct. The major drawback of using individual species as NAAQS indicators is the lack of  
12 routinely available measurement techniques and an associated resource burden even if adequate  
13 techniques were available. Currently, technology for measuring true NO<sub>2</sub>, HNO<sub>3</sub>, and PAN  
14 generally is not available for routine network applications. In addition to this practical  
15 consideration, there is another important reason for using the aggregated transference ratios.  
16 Because the standard, and the explicit Clean Air Act authority, is based on ambient air there must  
17 be an effective link connecting deposition and concentrations of the criteria pollutants in the  
18 ambient air. The use of individual species conceptually allows for a more physically meaningful  
19 approach to characterize and calculate deposition. However, the transference ratios also enable  
20 incorporation of the contributions of wet precipitation in the ambient air indicators. There is no  
21 practical alternative that allows for the disentangling of wet deposition as a function of ambient  
22 air concentrations as that relationship is best addressed through the coupling of numerous  
23 meteorological and chemical processes imbedded in the air quality modeling platform. One  
24 could consider wet precipitation as a separate parameter and isolate dry precipitation in the AAPI  
25 equation. But doing so would lose the important connection between wet precipitation of  
26 nitrogen and the same emission sources responsible for dry deposition.

27  
28            Finally, one might advocate for direct measurements of dry deposition of individual NO<sub>y</sub>  
29 species. Again, technologies simply are not ready for consideration in routine network  
30 applications. And, it is arguably practical to model dry deposition even if direct dry deposition  
31 measurement technologies were available. One reason for this is that there is significant spatial

1 heterogeneity in the factors (vegetation and surface type, micrometeorology) that define  
2 deposition velocity. Consequently, direct dry deposition measurements would have limited  
3 spatial representativeness in comparison to ambient air observations. The model conceptually  
4 accounts for the spatial variance, at the level of horizontal grid cell resolution, of the factors  
5 defining deposition velocity on a species by species basis. However, one also could reason that a  
6 well placed direct measurement of dry deposition is more realistic than a modeled result that  
7 relies on numerous assumptions. The development of technologies to measure direct dry  
8 deposition will benefit the diagnosis and improvement of process formulations in models.  
9

10 Consideration has been given to change the atmospheric indicator from  $\text{NO}_Y$  to total  
11 nitrate (the sum of nitric acid and particulate nitrate). The rationale for that approach is that a  
12 larger fraction of the deposited  $\text{NO}_Y$  is accounted for by total nitrate, which currently is  
13 measured in CASTNET with high confidence. One can reason adequately that nitrate may  
14 correlate well with total oxidized nitrogen deposition relative to  $\text{NO}_Y$  (as discussed in Chapter 4),  
15 given the inherent noise associated with variable contributions of low deposition velocity species  
16 of ambient level significance (e.g.,  $\text{NO}_2$ ). The disadvantages of using total nitrate as an  
17 indicator are that significant ambient mass with the potential for deposition is not captured, and  
18  $\text{NO}_Y$  is a preferred measurement for model evaluation and accountability purposes.  
19 Accountability refers to assessing if emissions reductions have the intended consequences on  
20 ambient air and deposition levels in the context of, “Aare our emissions reductions strategies  
21 working as planned?” All three of these concerns relate to the benefit of closing mass balances in  
22 whatever environmental medium is being characterized. In addition, the use of nitrate alone  
23 would create an increased distancing from the listed criteria pollutant oxides of nitrogen as  $\text{NO}_Y$   
24 does include NO and  $\text{NO}_2$ .

## 25 26 **8.2 Reactive Oxidized Nitrogen and Sulfur Species.**

27  
28  *$\text{NO}_Y$  species.* Air quality models and deposition models that use direct observations calculate  
29 ‘deposition on a species by species basis to account for differences in deposition velocities.  
30 Consequently, the relative fractional contributions of individual  $\text{NO}_Y$  or  $\text{SO}_X$  species to  
31 deposition or concentration is influenced by the differences in species deposition velocities. For

1 example, nitric acid with a high deposition velocity would exhibit a larger relative contribution  
2 to overall deposition compared to ambient concentrations in a particular area (Figure 8-1). The  
3 dominant ambient air NO<sub>Y</sub> species are NO, NO<sub>2</sub>, HNO<sub>3</sub>, P-NO<sub>3</sub> and PAN. Near source urban  
4 environments typically have a relatively higher fraction of NO<sub>x</sub> compared to the products of  
5 NO<sub>x</sub> reactions, nitrates and PAN, which are relatively more dominant in rural locations (Figures  
6 8-2 – 8-5).

7  
8 The differences in the relative patterns between ambient air and deposition on a species  
9 by species basis illustrate a number of challenges and considerations in developing a monitoring  
10 strategy. It is clear in the Adirondacks and Shenandoah areas that nitric acid is the most  
11 dominant contributing species from a deposition perspective (Figure 8-1), with significant  
12 contributions from particulate nitrate, PAN and NO<sub>2</sub>. The original source of emissions (NO  
13 accounts for 90-95% of all emitted NO<sub>x</sub>) provides very small (< 5%) contributions to ambient air  
14 and virtually nothing to deposition. The combination of nitric acid and particulate nitrate  
15 consistently contribute greater than 50% of the oxidized nitrogen dry deposition load, whereas  
16 PAN and NO<sub>2</sub> contribute roughly 15-25% of the deposition load. These broad summary  
17 statements speak to some of the monitoring considerations addressed earlier, particularly the case  
18 for monitoring for total nitrate. However, caution should be exercised when considering not  
19 measuring a considerable fraction of the ambient NO<sub>Y</sub> burden reflected in NO<sub>2</sub> and PAN.  
20 Characterization of NO<sub>2</sub> deposition is an area requiring further refinement especially considering  
21 that NO<sub>2</sub> is a significant component of total oxidized nitrogen. Zhang et al. (2005) suggest that  
22 NO<sub>2</sub> contributes up to 36% of dry NO<sub>Y</sub> deposition in rural Eastern Canadian locations, and  
23 suggest, based on observational evidence (Figure 8-2), that in some locations NO<sub>2</sub> deposition  
24 may be similar to nitric acid contributions.

25  
26 A sampling of co-located NO<sub>Y</sub> species observations in rural Eastern Canada (Figure 8-2),  
27 particularly in Egbert, Ontario, illustrates the concern of the general assumption that NO<sub>2</sub> may  
28 not contribute significantly to NO<sub>Y</sub> deposition in rural locations. While it may be true that in  
29 general NO<sub>2</sub> is of less of concern in rural areas relative to urban areas, that does not dismiss the  
30 potential for significant misrepresentation of total nitrogen budget in certain rural locations.

31

1 The results also raise the question of potential efficiencies gained from not cycling  
2 between NO and NO<sub>Y</sub> analyses, which is the standard configuration in commercial NO<sub>Y</sub>  
3 instruments, acknowledging the limited use of NO data in rural, acid sensitive environments  
4 (note that NO present in the air would still be captured in the NO<sub>Y</sub> measurement).  
5

6 These examples are used to support the rationalization of using NO<sub>Y</sub> as an appropriate  
7 atmospheric indicator in applying the AAPI. However, while it may be required to measure  
8 NO<sub>Y</sub> for explicit AAPI calculations to determine compliance with a NO<sub>x</sub>/SO<sub>x</sub> standard, there  
9 should be additional measurements of true NO<sub>2</sub>, HNO<sub>3</sub>, p-NO<sub>3</sub> and PAN to allow for diagnostic  
10 evaluations of both air quality models and the NO<sub>Y</sub> measurement itself. This recommendation  
11 would leverage existing CASTNET filter pack (FP) observations necessary to capture particulate  
12 sulfate (discussed below) and therefore require the addition of true NO<sub>2</sub> measurements and  
13 periodic sampling for PAN.  
14

15 ***Sulfur Species.*** Although sulfur dioxide and particulate sulfate contribute approximately 60 and  
16 40 %, respectively, to ambient SO<sub>x</sub> concentrations, sulfur dioxide is the dominant contributor to  
17 SO<sub>x</sub> deposition (Figure 8-6), which is consistent with CASTNET observational studies (Sickles  
18 and Shadwick, 2007). Measurement technology issues are not as complex for SO<sub>x</sub> as they are  
19 for NO<sub>Y</sub> and individual NO<sub>Y</sub> species. Issues related to particle size fraction and averaging  
20 period for SO<sub>x</sub> are discussed below.  
21

### 22 ***Reduced Nitrogen.***

23

24 The AAPI does not include reduced nitrogen (ammonia gas and ammonium ion) as an  
25 ambient air indicator. However, reduced nitrogen deposition is an explicit AAPI component  
26 which is estimated through air quality modeling. As discussed in the Chapter 7, characterization  
27 of reduced nitrogen deposition processes is an active developmental area which would benefit  
28 markedly from NH<sub>x</sub> measurements in order to assess modeled predictions of ambient patterns of  
29 ammonia and ammonium. This need for monitoring ammonia in rural environments is further  
30 supported by emerging evidence that ammonia acts as a regionally dispersed species based on  
31 the inclusion of ammonia bi-directional flux in CMAQ simulations. (Dennis et al., 2010).

1 Monitoring method approaches under consideration for routine application typically are limited  
2 to time averaged filter and denuder technologies, including passive sampling approaches.

3  
4 **8.3 What measurements would be used to characterize NO<sub>y</sub> and SO<sub>x</sub> ambient air**  
5 **concentrations for the purposes of the AAPI based standard?**

6  
7 Ambient NO<sub>y</sub>, SO<sub>2</sub> and particulate sulfate (SO<sub>4</sub>) concentrations would be used as the  
8 indicators in determining compliance with the standard. All of these indicators are measured in  
9 different places within the current routine monitoring networks (section 3.2). However, there are  
10 issues requiring resolution associated with Federal Reference or Equivalency Measurement  
11 (FRM/FEM) status of measurement techniques that to date have served as supplemental  
12 information, which will require resolution. A FRM for SO<sub>2</sub> exists, but not for NO<sub>y</sub> or SO<sub>4</sub>. Only  
13 recently have NO<sub>y</sub> measurements, which historically were viewed as research venue  
14 measurements, been incorporated as “routine” observations, partly as a result of the NCore  
15 program. Acquiring FRM status may require better characterization of the conversion  
16 efficiencies, mass loss and clear guidance on operating and siting procedures. Particulate sulfate  
17 has been measured for several years in the IMPROVE, CASTNET and EPA CSN networks. The  
18 nation has over 500 24-hour average, every third day sulfate measurements produced by the  
19 PM<sub>2.5</sub> speciation networks (IMPROVE and EPA CSN) and nearly 80 CASTNET sites that  
20 provide continuous weekly average samples of sulfate with an open inlet accommodating all  
21 particle sizes. With minor exceptions, the PM<sub>2.5</sub> fraction generally accounts for over 80% of the  
22 ambient sulfate mass. Unfortunately, as particle size diameters increase beyond 2.5 μ,  
23 gravitational settling imparts greater influence resulting in substantially enhanced deposition  
24 velocities. Consequently, the sulfate mass in size fractions greater than 2.5 μ potentially  
25 provides correspondingly greater contribution to as much as 50% of dry sulfate deposition in  
26 certain locations (Grantz et al., 2003).

27  
28 Sample collection period is not an issue for gaseous measurements of NO<sub>y</sub> and SO<sub>2</sub> that  
29 operate continuously. However, consideration should be given to using the CASTNET FP for  
30 SO<sub>2</sub> measurements as a resource saving option, assuming the FPs will be used for particulate  
31 sulfate. However, the availability of highly time resolved data will support the continual

1 evaluation of SO<sub>2</sub> and sulfate balance in air quality modeling systems which is a critical  
2 underpinning for both human and ecosystem health assessments. Some concerns have been  
3 raised about the possibility of exclusion of coarse particles from NO<sub>Y</sub> samplers operating at low  
4 flow conditions as well as potential difficulties of reducing organically bound and mineralized  
5 nitrate. Insight into conversion and capture efficiency characteristics will be advanced both by  
6 research catalyzed by the need to support this standard and through ongoing and future network  
7 operations.

8

#### 9 **8.4 What additional complementary measurements are recommended?**

10

11 We recommend that there be 3-4 locations nationally, in airsheds with different  
12 atmospheric chemistries, that sample not only for the NAAQS indicator NO<sub>Y</sub> but for the suite of  
13 major NO<sub>Y</sub> species as well; HNO<sub>3</sub>, p-NO<sub>3</sub>, PAN, true NO<sub>2</sub>, and NO as discussed earlier. Not  
14 only is this important from a modeling and process diagnosis perspective, but it is especially  
15 useful in the introduction of new measurements that have a limited track record to provide  
16 insight into instrument performance. In the case of NO<sub>Y</sub>, it is even more relevant since there  
17 effectively are no standards that explicitly challenge instrument accuracy given the highly  
18 variable nature of NO<sub>Y</sub> species distribution and the instability associated with mixing NO<sub>Y</sub> gases.  
19 This quality assurance issue is analogous to PM<sub>2.5</sub> where aerosol standards are not available and  
20 measurement accuracy is judged against periodic challenges relative to a “gold standard”  
21 instrument. Reduced nitrogen measurements of ammonia and ammonium ion are recommended  
22 at all locations with FRM/FEM instruments based on the need to support the AAPI as discussed  
23 above.

24

#### 25 **8.5 What sampling frequency would be required?**

26

27 The averaging time for the standard is likely to be an annual average, perhaps based on 3-  
28 5 years of data collection to minimize the influence of interannual variability in meteorology,  
29 especially precipitation.. Conceptually, extended sampling periods no longer than one year  
30 would be adequate for the specific purposes of comparison to a standard. However, there are  
31 significant peripheral benefits relevant to improving the scientific foundation for subsequent

1 reviews and a variety of related air quality and deposition assessments to be gleaned from more  
2 highly time resolved data. In particular, the critical role of air quality models in deposition  
3 assessments implies value to be derived from measurements that support model evaluation and  
4 improvement. Many of the monitoring approaches that are used throughout the nation sample  
5 (or at least report out) on daily (PM<sub>2.5</sub> chemical speciation), weekly (CASTNET) and hourly (all  
6 inorganic gases) periods. There is a tradeoff to consider in sampling period design. For  
7 example, the weekly CASTNET collection scheme covers all time periods throughout a year, but  
8 only provides weekly resolution that misses key temporal and episodic features valuable for  
9 diagnosing model behavior. The every third day, 24-hour sampling scheme used in IMPROVE  
10 and EPA speciation monitoring does provide more information for a specific day of interest yet  
11 misses 2/3 of all sampling periods. The missing sampling period generally is not a concern when  
12 aggregating upward to a longer term average value as the sample number adequately represents  
13 an aggregated mean value. Additionally, there is a benefit to leveraging existing networks which  
14 should be considered in sampling frequency recommendations. A possible starting point would  
15 be to assume gaseous oxidized species, NO<sub>y</sub> and SO<sub>2</sub>, are run continually all year reporting  
16 values every hour, consistent with current routine network operations. Sulfate sampling periods  
17 should coincide with either the chemical speciation network schedules or with CASTNET.  
18 There are advantages to coordinating with either network. Ammonia gas and ammonium ion  
19 present challenges in that they are not routinely sampled and analyzed for, and the combined  
20 quantity, NH<sub>x</sub> is of interest. Because NH<sub>x</sub> is of interest, some of the problems of volatile  
21 ammonia loss from filters may be mitigated. However, for model diagnostic purposes,  
22 delineation of both species at the highest temporal resolution is preferred.

23

## 24 **8.6 What are the spatial scale issues associated with monitoring for compliance, and** 25 **how should these be addressed?**

26

27 The current observation network for NO<sub>y</sub>, NH<sub>x</sub> and SO<sub>x</sub> is very modest and includes a  
28 monitoring network infrastructure that is largely population oriented with the exception of  
29 CASTNET and IMPROVE. While there is platform and access infrastructure support provided  
30 by CASTNET, NADP and IMPROVE, those locations by themselves are not likely to provide  
31 the needed spatial coverage to address acid sensitive watersheds across the United States.

1 Ambient monitoring at every watershed will not be required given the reality of resource  
2 constraints and the relative spatial homogeneity of air concentrations that are averaged over  
3 annual time periods and within ‘acid sensitive’ areas. The spatial monitoring requirements will  
4 be associated with the determination of acid sensitive areas, which is discussed at length in  
5 Chapter 5. The number of sites per area will be addressed in rule development and general  
6 guidance based on an understanding of the spatial variability of  $\text{NO}_Y$ ,  $\text{NH}_X$ , sulfate and  $\text{SO}_2$   
7 combined with resource allocations will help inform those decisions.

8  
9 Critical load models applied for the purposes of this standard would be based on annual  
10 averages, which effectively serves to dampen much of the spatial variability. Furthermore, the  
11 development of an area-wide depositional load tradeoff curve implies focus on region wide  
12 characterization. Toward that end, CMAQ concentration fields will provide insight into the  
13 likely spatial representativeness of monitors leading to efficient application of monitoring  
14 resources. For example, the CMAQ based spatial coefficient of variation (standard  
15 deviation/mean) of oxidized nitrogen in the Adirondacks was 1.46%. Improved dry deposition  
16 estimates will result from enhancements of ambient monitoring addressing the N/S secondary  
17 standards as each additional location could serve a similar role that existing CASTNET sites  
18 provide in estimating dry deposition.

## 19 20 **8.7 What specific monitoring methods would be used?**

21  
22 Federal reference and/or equivalent methods (FRM/FEM) are presently available only for  
23  $\text{SO}_2$ . Particulate  $\text{SO}_4$  is measured at over 500 sites nationally, and there is a general consensus  
24 that methods available are reliable and provide consistent data.  $\text{NO}_Y$  measurement is in a  
25 transition period from largely being viewed as a research level measurement to now being  
26 deployed as a routine measurement in EPA’s national 75 site NCORE network. The general  
27 consensus on  $\text{NO}_Y$  measurement is that the methodology is sound and applicable for  
28 routine/regulatory use, but there does not exist a well defined understanding of the quality of  
29  $\text{NO}_Y$  data. Inorganic dry nitrate (nitric acid and particulate nitrate) is measured routinely in the  
30 CASTNET network with filter packs (FP).

1           One of the challenges associated with specifying performance attributes for p-SO<sub>4</sub> and  
2 NO<sub>Y</sub> is the lack of specific challenge standards. For example, instruments measuring discrete  
3 gases such as ozone or nitrogen oxide can be challenged by comparing an instrument's reading  
4 when measuring known concentrations of gases which are readily provided for single gas  
5 concentrations. Particle standards are not available. NO<sub>Y</sub> performance typically is challenged by  
6 known mixtures of NO<sub>2</sub>, and occasionally with N-propyl nitrate, which only addresses part of the  
7 spectrum of nitrogen species in an NO<sub>Y</sub> mix. Consequently, instrument performance in EPA's  
8 national networks for aerosol mass is quantified in terms of bias and precision relative to a co-  
9 located "performance evaluation" instrument. There is no comparable program in place for p-  
10 SO<sub>4</sub> or NO<sub>Y</sub>.

11  
12           **p-SO<sub>4</sub>.** The routinely operating methodology for particulate sulfate (p-SO<sub>4</sub>) is based on  
13 an integrated (i.e., time averaged over several hours or days) sample collection on a Teflon filter  
14 followed by ion chromatography (IC) detection in the laboratory. Two major variations of this  
15 approach are applied in the PM<sub>2.5</sub> speciation (exclusion of particles larger than 2.5 μ and 24-hour  
16 collection typically every third day) and CASTNET (weekly average integrated sampling all year  
17 with an open inlet to include all size fractions). There are additional variations related to inlet  
18 design and flow characteristics of PM<sub>2.5</sub> speciation samplers in which two designs are prevalent  
19 in the networks: (IMPROVE and EPA CSN SASS samplers). These variations are considered  
20 minor as sulfate species (dominated by ammonium sulfate) typically are not subject to major  
21 sampling artifacts associated with volatilization or condensation. The difference in inlets (open  
22 vs. 2.5 μ) is perceived by some as not an issue of concern as 80 - 90 % of the PM sulfate mass is  
23 distributed in size fractions less than 2.5 μ. However, the higher deposition velocities associated  
24 with larger diameter particles argue for including all size fractions as discussed above.  
25 Continuously operating in-situ sulfate instruments that allow for hourly, or less, data reporting  
26 are available. However, the limited deployment (less than 20 sites nationally) of these  
27 instruments combined with the 2.5 μ inlet cutoff configuration preclude consideration at this  
28 time.

29  
30           The CASTNET FP offers three important attributes: a history of high quality data,  
31 existing infrastructure and network to build on and an open inlet to capture the full range of

1 particle diameters. EPA intends to develop FRM status for this method. A significant  
2 additional advantages of using the FP method will be the availability of important co-measured  
3 species (e.g., SO<sub>2</sub>, total nitrate, ammonium). While EPA will expedite the certification process  
4 for the CASTNET FP, in the future consideration should be given to other available methods to  
5 more efficiently leverage network assets. For example, the SASS sampler potentially would  
6 accommodate ammonia gas and ammonium ion measurements, as well as other standard  
7 chemical speciation parameters depending on the configuration of this multi channel system.  
8 Continuous sulfate measurements would be extremely useful for model evaluation, especially  
9 considering the availability of continuous SO<sub>2</sub> data that would be required as part of the NAAQS  
10 indicators. A performance based approach to meet equivalency requirements, given the variety  
11 of sulfate measurement approaches and well vetted and accurate analytical procedures.

12  
13 **SO<sub>2</sub>.** Sulfur dioxide is a NAAQS pollutant and a FRM is available. See 75 FR at 35554-  
14 56 and 35593-95 (June 22, 2010) (adopting a second FRM for SO<sub>2</sub>). As part of the NCore  
15 network development effort, trace gas SO<sub>2</sub> analyzers capable of sub ppb resolution became  
16 commercially available and are the preferred instruments for implementation in rural locations.  
17 As discussed above, the near continuous data output of gaseous analyzers is desired for  
18 peripheral support of model evaluation. Nevertheless, the convenience and resource savings  
19 associated with the CASTNET FP suggest that Federal Equivalency Method (FEM) status should  
20 be incorporated in concert with the sulfate certification process.

21  
22 **NO<sub>y</sub>.** In principle, measured NO<sub>y</sub> is based on catalytic conversion of all oxidized  
23 species to NO followed by chemiluminescence NO detection. While there are caveats associated  
24 with instrument conversion efficiency and possible inlet losses, the technique is considered  
25 adequate and routinely operational. Approximately 25 sites (out of a planned 75) in EPA's  
26 NCore network are operating NO<sub>y</sub> instruments, and an additional 5-10 sites are operated in  
27 SEARCH, CASTNET and other programs. NO<sub>y</sub> measurements are nearly continuous, reporting  
28 at hourly intervals providing far greater temporal information compared to filter or denuder  
29 based methods. FRM certification for NO<sub>y</sub> presents more considerable challenges given the  
30 limited history of routinely operating instruments. The process EPA is pursuing for certification  
31 status for NO<sub>y</sub> will be addressed in the final PA.

1 **8.8 REFERENCES**

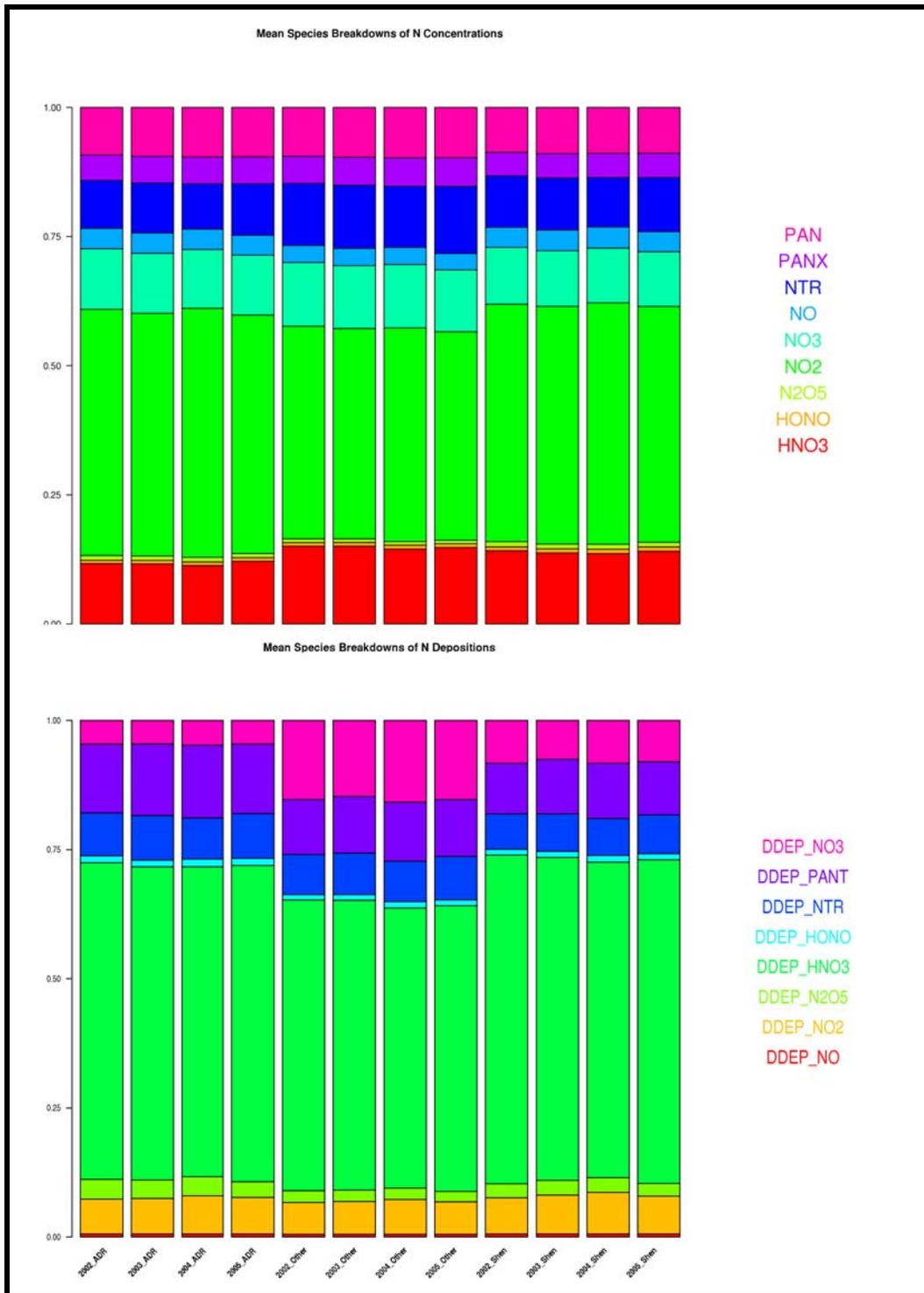
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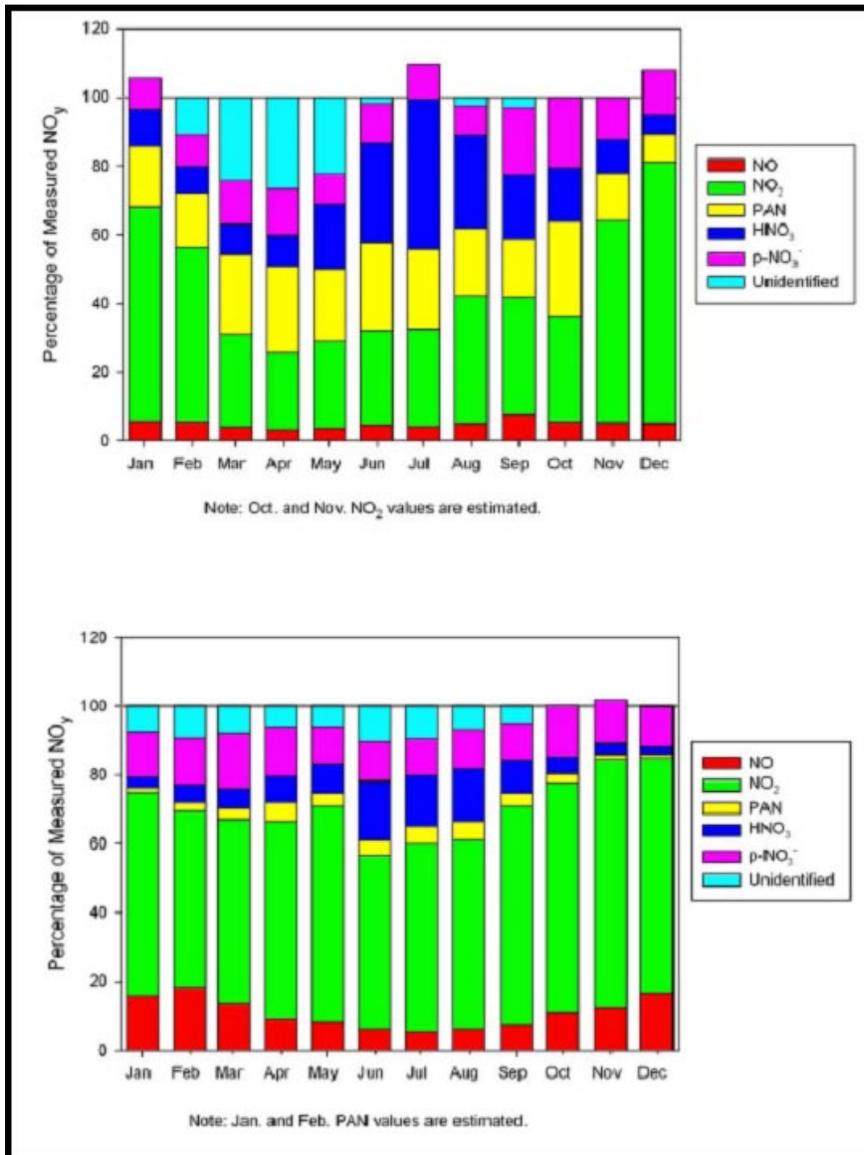
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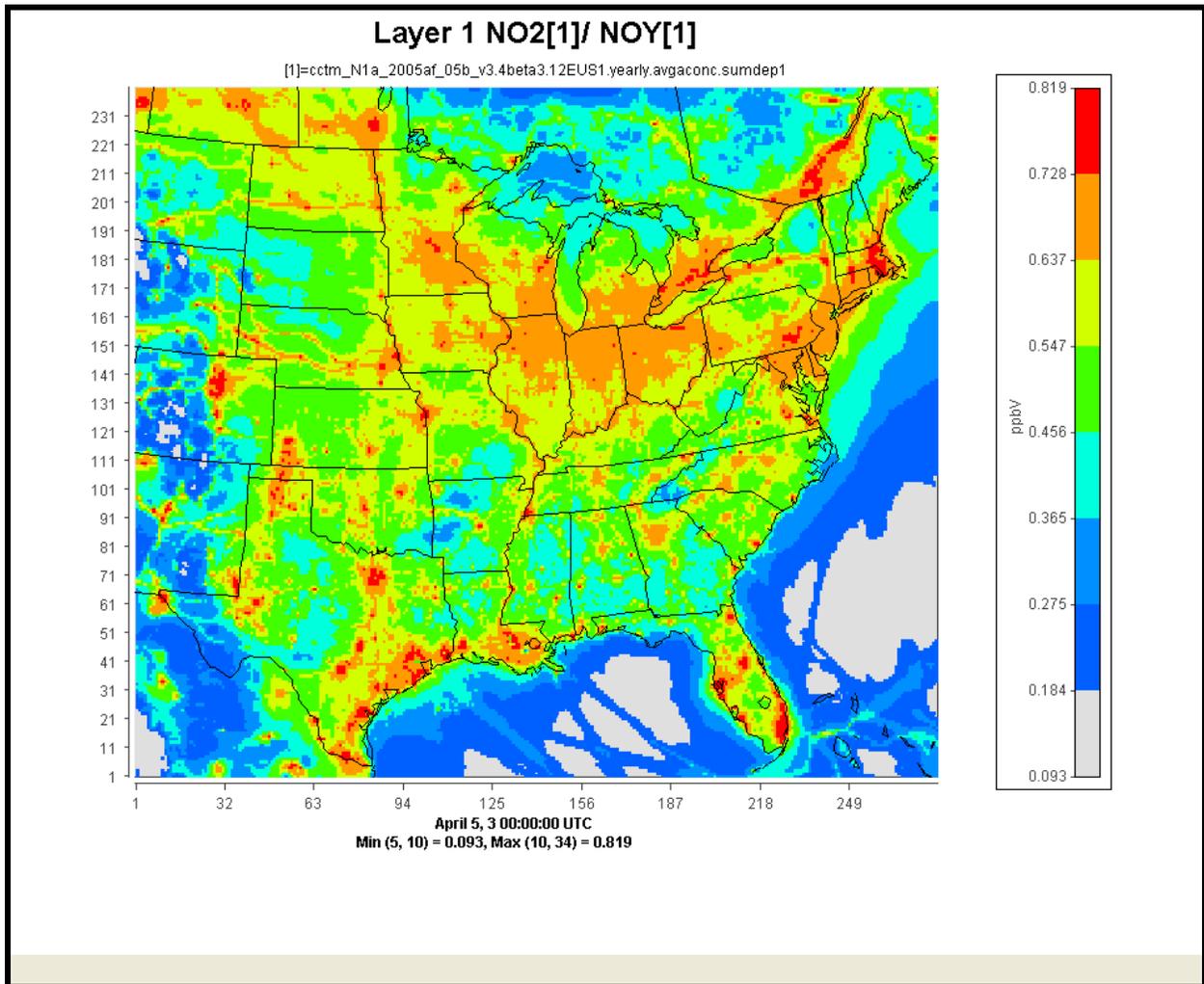
2 **Figure 8-1 Annual 2002 – 2004 CMAQ derived annual average fraction of ambient**  
 3 **concentrations (above ) and deposition (below) of individual NO<sub>y</sub> species**  
 4 **delineated by the Adirondack and Shenandoah case study areas and the**  
 5 **remainder of the Eastern U.S. domain.**



2

3 **Figure 8-2** Examples of the Relative Abundance of Several NO<sub>y</sub> Species Measured at Two  
 4 Rural Southeastern Canadian Sites as a Fraction of the Total Measured NO<sub>y</sub>  
 5 Concentration -- Kejimkujik, NS, (top) and Egbert, ON, (bottom) during 2003.  
 6 Although both sites are in rural locations, the Kejimkujik, NS site represents more  
 7 aged air masses as it lies considerably further downwind from major sources of  
 8 NO<sub>x</sub> relative to the Egbert site. (*Source: NARSTO, 2010*)

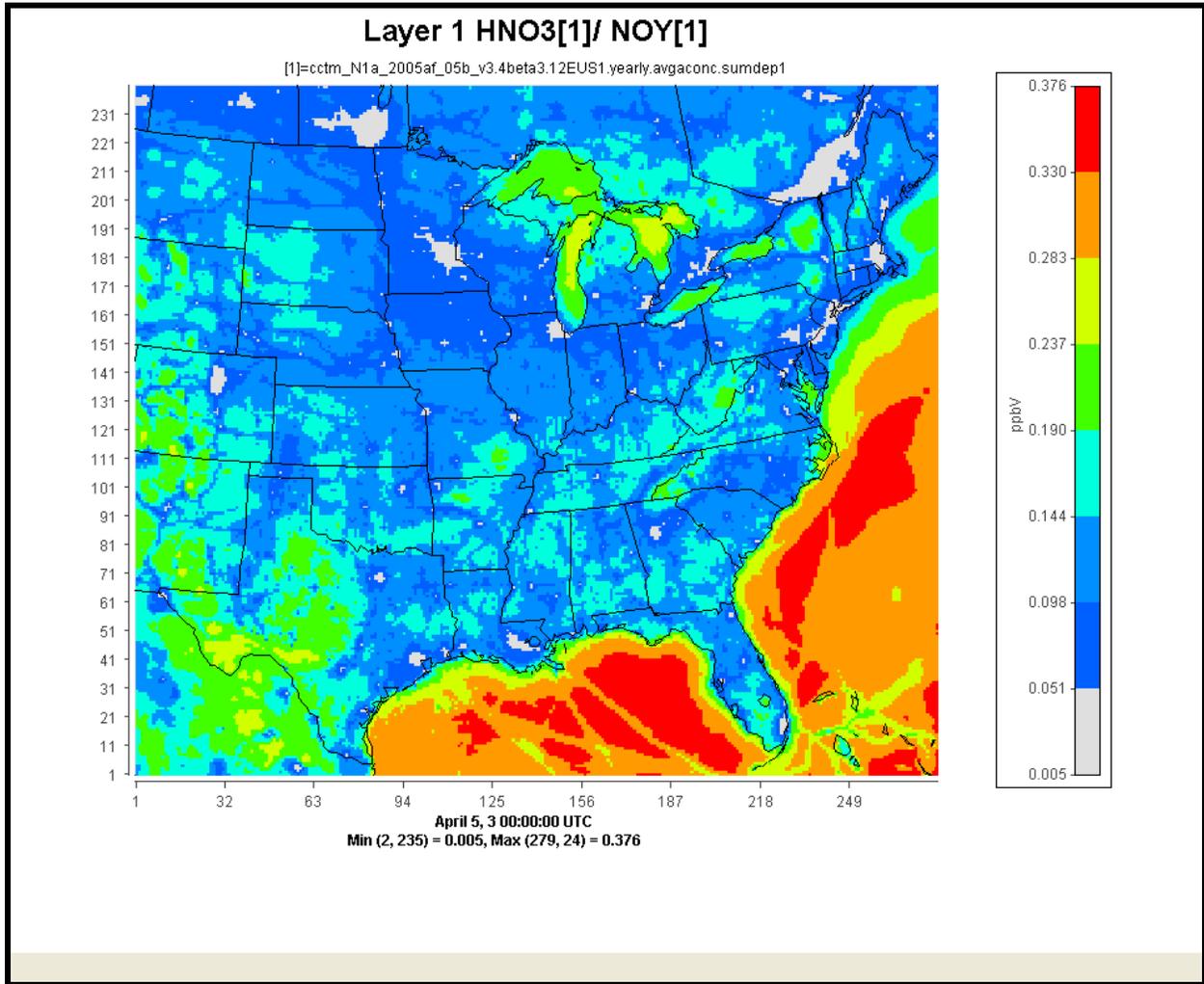
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3 **Figure 8-3 Annual average fraction of NO<sub>y</sub> ambient air contributed by NO<sub>2</sub> based on**  
4 **2005 CMAQ Eastern U.S. simulation at 12 km grid cell resolution.**

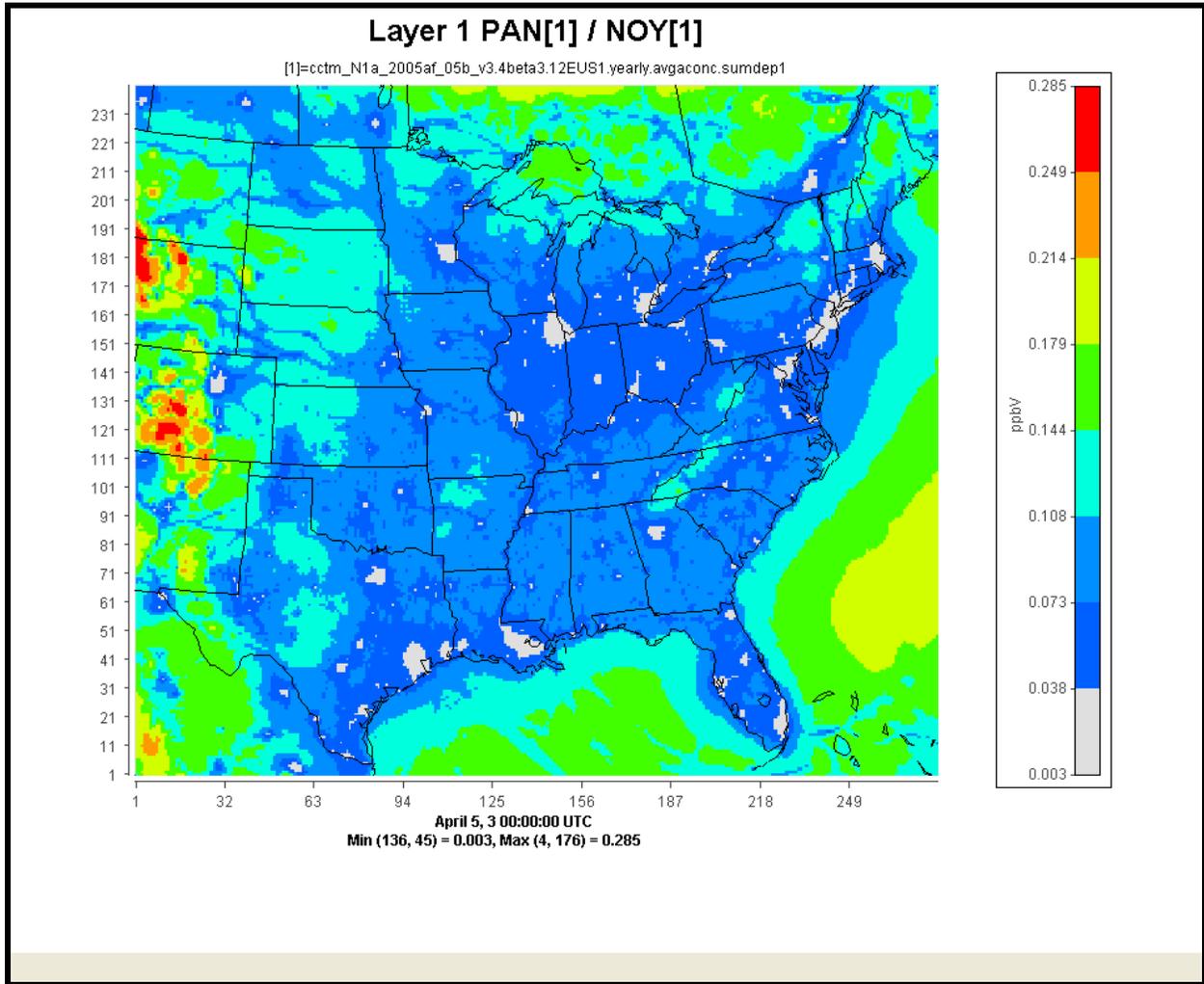
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3 **Figure 8-4 Annual average fraction of NO<sub>y</sub> ambient air contributed by HNO<sub>3</sub> based on**  
4 **2005 CMAQ Eastern U.S. simulation at 12 km grid cell resolution.**

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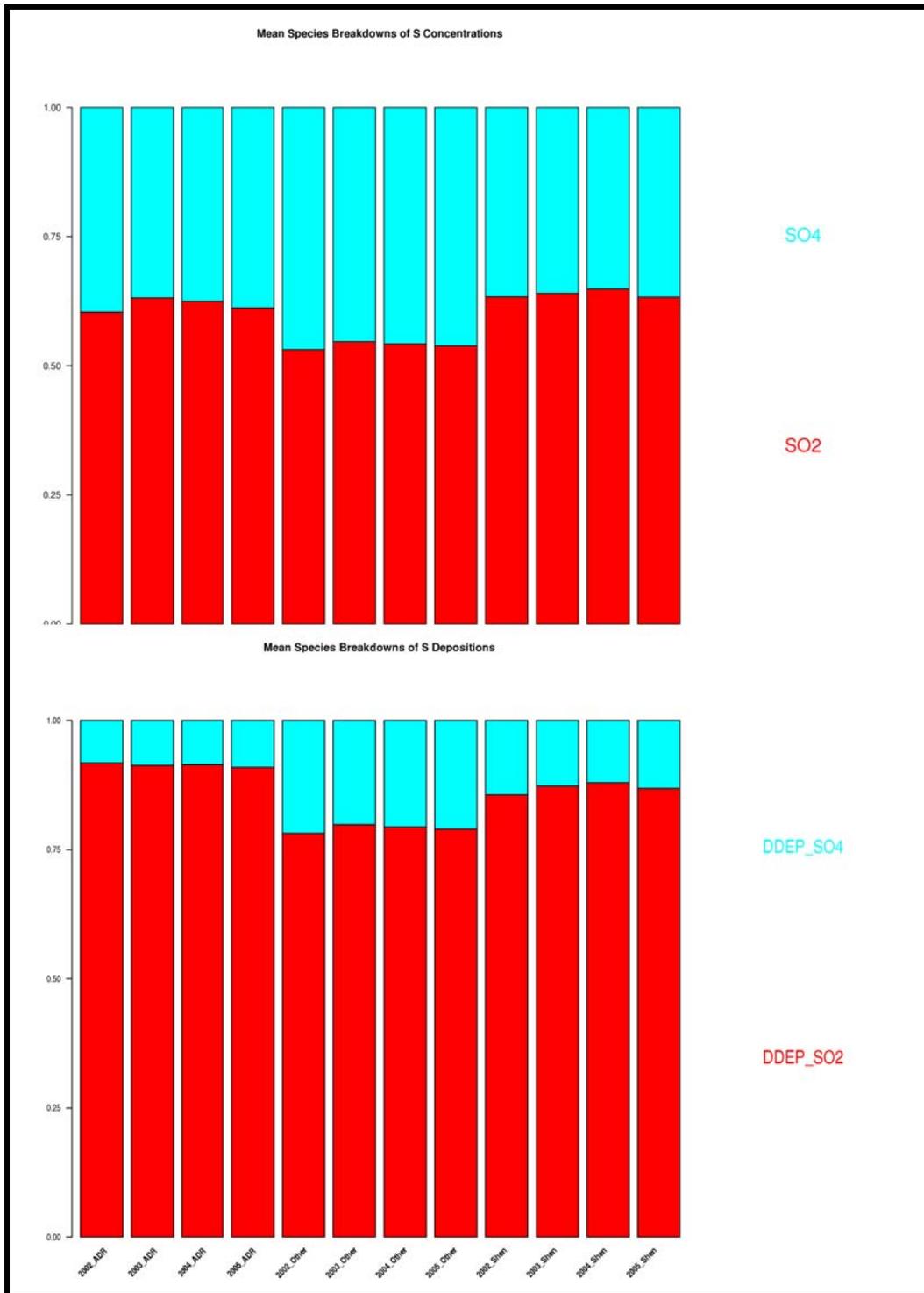


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3 **Figure 8-5 Annual average fraction of NO<sub>y</sub> ambient air contributed by PAN based on**  
4 **2005 CMAQ Eastern U.S. simulation at 12 km grid cell resolution.**

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 2 **Figure 8-6 Annual 2002 – 2004 CMAQ derived annual average fraction of ambient**  
 3 **concentrations (above ) and deposition (below) of individual SO<sub>x</sub> species**  
 4 **delineated by the Adirondack and Shenandoah case study areas and the**  
 5 **remainder of the Eastern U.S. domain.**  
 6

## 9 INITIAL CONCLUSIONS

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2  
3 Staff initial conclusions on the elements of the secondary NO<sub>x</sub> and SO<sub>x</sub> standards for the  
4 Administrator's consideration in making decisions on the secondary NO<sub>x</sub> and SO<sub>x</sub> standards are  
5 summarized below, together with supporting conclusions from previous chapters. We recognize  
6 that selecting from among alternative policy options will necessarily reflect consideration of  
7 qualitative and quantitative uncertainties inherent in the relevant evidence and in the assumptions  
8 of the quantitative exposure and risk assessments. Any such standard should protect public  
9 welfare from any known or anticipated adverse effects associated with the presence of the  
10 pollutant(s) in the ambient air, "whether caused by transformation, conversion, or combination  
11 with other air pollutants." CAA § 302(h). In providing these options for consideration, we are  
12 mindful that the Act requires standards that, in the judgment of the Administrator, are requisite to  
13 protect public welfare. The standards are to be neither more nor less stringent than necessary.  
14 Our focus in this review on ecosystems that are both sensitive to acidification and responsive to  
15 atmospheric acid deposition is intended to ensure that the resulting standards are appropriately  
16 protective and not more protective than necessary in ecosystems that are not adversely affected  
17 by acid deposition.

18 To evaluate whether the current secondary NAAQS is adequate or whether consideration  
19 of revisions is appropriate, the conclusions and options for the Administrator to consider in this  
20 review are based on effects-, exposure- and risk-based considerations. The exposure and risk  
21 assessments reflect the availability of new tools, assessment methods, and a larger and more  
22 diverse body of evidence than was available in the last reviews. We have taken a weight of  
23 evidence approach that evaluates information across the variety of research areas described in the  
24 ISA and in addition includes assessments of air quality, exposures, and qualitative and  
25 quantitative risks associated with alternative air quality scenarios.

26 Staff notes that since the last review, additional policy-relevant developments have  
27 occurred that may also warrant consideration by the Administrator when making decisions about  
28 what is requisite to protect public welfare. The NRC report (described in Chapter 5) states:  
29 "Whatever the reason that led EPA to use identical primary and secondary NAAQS in the past, it  
30 is becoming increasingly evident that a new approach will be needed in the future. There is

1 growing evidence that the current forms of the NAAQS are not providing adequate protection to  
2 sensitive ecosystems and crops” (NRC, 2004).

3 The last review raised the following key issues as a rationale for not setting a separate  
4 standard for NO<sub>x</sub> to protect against acidification and nutrient enrichment effects in sensitive  
5 ecosystems:

6 1) Lack of enough consistent information to support a revision of the current secondary  
7 standard to protect these aquatic systems.

8 2) Lack of adequate quantitative evidence on the relationship between deposition rates  
9 and environmental impacts

10 3) Significant uncertainties with regard to the long-term role of nitrogen deposition in  
11 surface water acidity and with regard to the quantification of the magnitude and timing of the  
12 relationship between atmospheric deposition and the appearance of nitrogen in surface water.

13 In this current review, staff concludes that important new information has become  
14 available since the last review that supports revising the current NO<sub>x</sub> and SO<sub>x</sub> standards.  
15 Specifically, the ISA has concluded that there are causal relationships between NO<sub>x</sub> and SO<sub>x</sub>  
16 acidifying deposition and effects on aquatic and terrestrial ecosystems, and the ISA and REA  
17 provide substantial quantitative evidence of effects occurring in locations that meet the current  
18 NO<sub>2</sub> and SO<sub>2</sub> standards. In addition, substantial new information, based on observational data  
19 and rigorous atmospheric modeling, has become available regarding the role of both nitrogen and  
20 sulfur deposition in acidification of sensitive water bodies. This information is sufficient to  
21 inform the development of revised secondary standards for NO<sub>x</sub> and SO<sub>x</sub> to protect against the  
22 effects of acidification. While there is also new information available on the role of nitrogen  
23 deposition on nutrient enrichment effects in terrestrial and aquatic ecosystems, and the ISA  
24 concludes there is a causal relationship between NO<sub>x</sub> and nutrient enrichment effects, for this  
25 draft policy assessment, staff have focused on aquatic acidification effects due to the  
26 substantially greater amount of information available to inform the development of secondary  
27 standards for those effects. This is consistent with the available science and data, and also with  
28 the recommendations of CASAC, which stated that “EPA Staff is advised to focus on an AAPI  
29 standard driven by aquatic effects concerns.” There is not sufficient information at this time to  
30 develop secondary standards directly focused on protection of sensitive terrestrial ecosystems  
31 from acidification or sensitive terrestrial and aquatic ecosystems from adverse effects from

1 nutrient enrichment. Establishing standards that are multipollutant and ecologically relevant is  
2 an inherently complex process. We note that the aquatic acidification based standards are an  
3 important step in providing additional protections for sensitive ecosystems. While they do not  
4 provide complete protection for all sensitive ecosystems against all adverse effects, they will  
5 likely result in reductions in  $\text{NO}_Y$  and  $\text{SO}_2$  across broad regions of the U.S., resulting in  
6 decreased deposition and related effects for both terrestrial and aquatic ecosystems across all  
7 types of effects (see Chapter 6 for a broader discussion of this issue).

8 Staff highlights the progress made in considering the joint nature of ecosystem responses  
9 to acidifying deposition of  $\text{NO}_X$  and  $\text{SO}_X$ , and notes that the ability to consider revisions to the  
10  $\text{NO}_X$  and  $\text{SO}_X$  secondary standards has been enhanced by our ability to consider a joint standard  
11 for  $\text{NO}_X$  and  $\text{SO}_X$  to protect against aquatic acidification effects. The development of an  
12 appropriate form of the standard linked to a common indicator of aquatic acidification, ANC, is  
13 also a significant step forward, as it allows for development of a standard for aquatic  
14 acidification designed to provide generally the same degree of protection across the country,  
15 while still reflecting the underlying variability in ecosystem sensitivity to acidifying  $\text{NO}_X$  and  
16  $\text{SO}_X$  deposition.

17

## 18 **9.1 CONSIDERATION OF ALTERNATIVE STANDARDS**

19

20 We begin by noting that the existing evidence continues to support existing  $\text{NO}_2$  and  $\text{SO}_2$   
21 standards to protect against adverse effects associated with direct exposure of vegetation to gas  
22 phase  $\text{NO}_X$  and  $\text{SO}_X$ . The ISA concluded that there was sufficient evidence to infer a causal  
23 relationship between exposure to  $\text{SO}_2$ ,  $\text{NO}$ ,  $\text{NO}_2$  and PAN and injury to vegetation. Additional  
24 research on acute foliar injury has been limited and there is no evidence to suggest foliar injury  
25 below the levels of the current secondary standards for  $\text{SO}_X$  and  $\text{NO}_X$ . There is sufficient  
26 evidence to suggest that the levels of the current standards are likely adequate to protect against  
27 direct phytotoxic effects. As such, staff concludes that retaining the existing  $\text{NO}_2$  and  $\text{SO}_2$   
28 standards to continue protection against these effects is appropriate. However, as discussed in  
29 Chapter 4, we also conclude that the existing secondary  $\text{NO}_X$  and  $\text{SO}_X$  standards are not adequate  
30 to provide protection of aquatic and terrestrial ecosystems against the effects of acidifying  
31 deposition. In response to this conclusion, staff considers a second overarching question:

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*What additional NO<sub>x</sub> and SO<sub>x</sub> standards are supported by the currently available scientific evidence and risk-based information, as reflected in the ISA and REA?*

To inform the answer to this overarching question, we have posed a series of more specific questions to aid in considering how the current NO<sub>x</sub> and SO<sub>x</sub> standards might be revised to provide requisite public welfare protection.

Chapter 5 provided a conceptual framework for a secondary standard that is designed to provide protection of ecosystems against the effects associated with deposition of ambient concentrations of NO<sub>x</sub> and SO<sub>x</sub>. Chapter 5 also provided a discussion of potential options for the elements of a standard based on that conceptual framework, with a focus on the form of the standards. While we recognize the potential for significant impacts of current levels of NO<sub>x</sub> and SO<sub>x</sub> on terrestrial ecosystems and the effect of current levels of NO<sub>x</sub> on nutrient enrichment in sensitive aquatic ecosystems, we conclude that the currently available information is insufficient to develop either individual or joint standards to protect against these effects. We note that development of a standard for protection against terrestrial acidification may be appropriate using the same structure as we are proposing for aquatic acidification, using the Bc:Al ratio as the ecological indicator. However, the data needed to parameterize the form of such a standard is not sufficient at this time. As a result, we conclude that the current state of knowledge supports a standard to protect against the adverse effects associated with acidification of aquatic ecosystems. Such a standard is likely to provide some level of protection against other endpoints associated with deposition of N and S, but is not likely to adequately protect all sensitive terrestrial ecosystems or all N nutrient sensitive aquatic ecosystems.

Building on the options discussed in Chapter 5, this section offers staff conclusions regarding the elements of the standard, including the indicators for NO<sub>x</sub> and SO<sub>x</sub>, the form of the standard, the averaging times, and presents for consideration options for target ANC levels associated with protection against specific ecological effects in aquatic ecosystems. Ultimately, the levels of AAPI considered by the Administrator should incorporate consideration of target levels of ANC, percent of waterbodies protected within defined spatial areas, and trajectories for ecosystem recovery, as well as uncertainties in the components of the AAPI. Associated with these elements of the AAPI are sets of NO<sub>y</sub>/(SO<sub>2</sub>+SO<sub>4</sub>) tradeoff curves which determine the

1 levels of ambient  $\text{NO}_y$  and  $\text{SO}_2+\text{SO}_4$  which will satisfy the level of the standard. The  
2 expression of these trade-off curves embodies the depositional load for a specified spatial area  
3 that is equivalent to the critical load for the waterbody in that area that represents a selected  
4 percentile (e.g. 95<sup>th</sup> percentile) of critical loads across waterbodies in the area, such that the  
5 selected percent of waterbodies in that area are expected to achieve an ANC at the target level.  
6 If a target load for a specific temporal period (e.g. by 2030) is evaluated instead of the critical  
7 load, then the depositional load represents the amount of deposition that is expected to achieve a  
8 target ANC value by a specific year for the selected percent of waterbodies in the area. The  
9 equivalent AAPI for an area can be calculated by inputting the values for each parameter of the  
10 AAPI equation for the selected percentile waterbody and the observed values of  $\text{NO}_y$  and  
11  $\text{SO}_2+\text{SO}_4$ .

12 These elements will be considered collectively in evaluating the protection from welfare  
13 effects associated with aquatic acidification afforded by alternative standards under  
14 consideration. In considering the currently available scientific and technical information, we  
15 consider both the information available in the last review and information that is newly available  
16 since the last review as assessed and presented in the ISA and RA prepared for this review (US  
17 EPA, 2008; US EPA, 2009).

18

### 19 **9.1.1. Indicators**

20

21 Staff concludes that the appropriate indicators for  $\text{NO}_x$  and  $\text{SO}_x$ , as described in detail in  
22 Chapter 5, are total  $\text{NO}_y$  and the sum of  $\text{SO}_2$  and  $\text{SO}_4$ , respectively. Total  $\text{NO}_y$  includes all  
23 nitrogen oxides, including e.g. total reactive oxidized atmospheric nitrogen, defined as  $\text{NO}_x$  ( $\text{NO}$   
24 and  $\text{NO}_2$ ) and all oxidized  $\text{NO}_x$  products:  $\text{NO}_y = \text{NO}_2 + \text{NO} + \text{HNO}_3 + \text{PAN} + 2\text{N}_2\text{O}_5 +$   
25  $\text{HONO} + \text{NO}_3 + \text{organic nitrates} + \text{particulate NO}_3$ . The sum of  $\text{SO}_2$  and  $\text{SO}_4$  constitutes  
26 virtually the entire ambient air sulfur budget and  $\text{SO}_2$  and  $\text{SO}_4$  are measured routinely in  
27 existing monitoring networks.

28

### 29 **9.1.2. Averaging times**

30

1 As noted in Chapter 2 and Chapter 5, episodes of acidification and chronic acidification  
2 levels are associated with deposition over longer periods of time due to the storage and release of  
3 deposited nitrogen and sulfur in soils, snow, and ice. As a result, while episodic acidification  
4 may occur on much shorter timeframes, e.g. days to weeks, the cause of these episodes is largely  
5 due to shifts in hydrological flow paths, and the impact of these episodes is still determined by  
6 long term deposition of  $\text{NO}_y$  and  $\text{SO}_2+\text{SO}_4$  and associated long-term ANC, and thus the  
7 appropriate averaging time for ambient  $\text{NO}_y$  and  $\text{SO}_2+\text{SO}_4$  will be longer term. The averaging  
8 time for ambient concentrations of  $\text{NO}_y$  and  $\text{SO}_2+\text{SO}_4$  should be reflective of the long-term  
9 cumulative nature of deposition. CASAC supports using a three to five year averaging period  
10 “to help smooth out the year-to-year climatic variation in air concentration and deposition  
11 estimates.” (CASAC, 2010)

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### 13 **9.1.3. Form**

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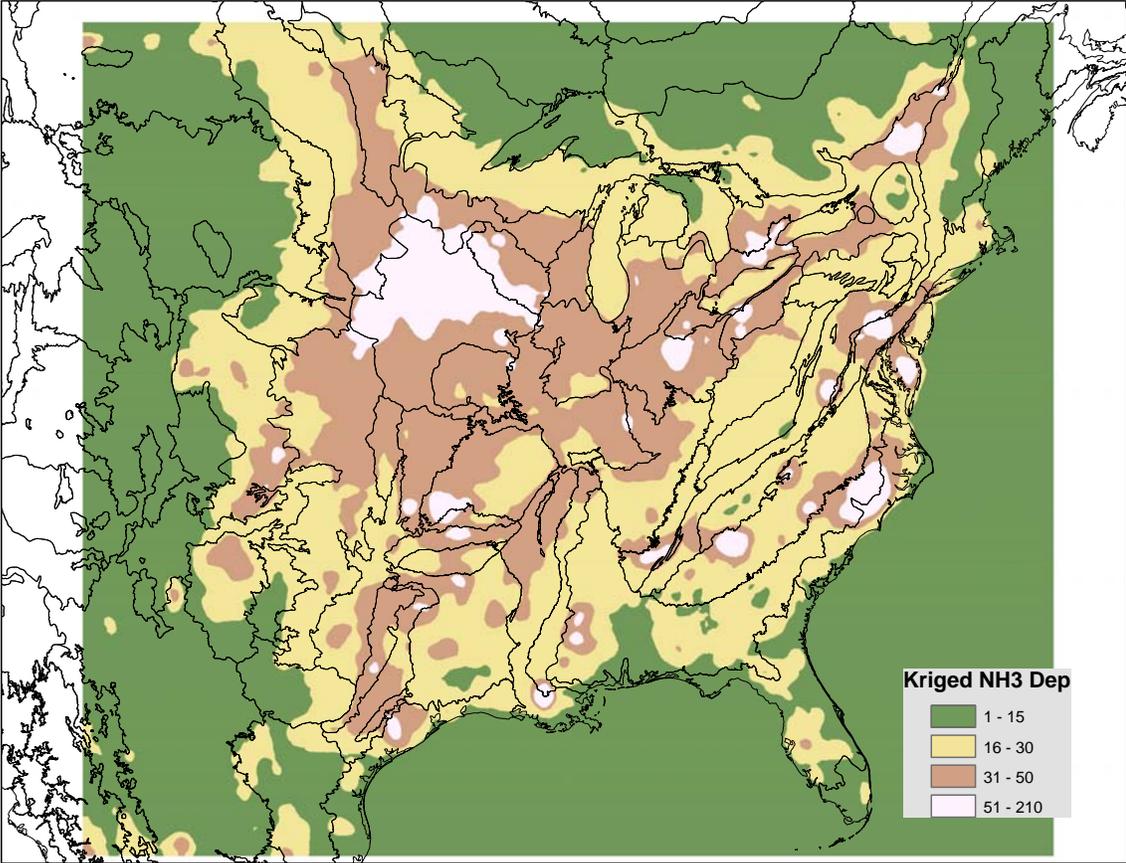
15 The “form” of a standard defines the air quality statistic that is to be compared to the  
16 level of the standard in determining whether an area attains the standard. As discussed  
17 extensively in chapter 5, staff concludes that the current forms of the  $\text{NO}_x$  and  $\text{SO}_x$  secondary  
18 standards are not appropriate for addressing ecosystem acidification effects, and also concludes  
19 that a form that combines  $\text{NO}_x$  and  $\text{SO}_x$  levels with information on ecosystem sensitivity and  
20 nitrogen retention and uptake is most appropriate to maximize the likelihood of protecting  
21 sensitive ecosystems from the effects of acidification, without requiring standards that are more  
22 than requisite to provide that protection. Specifically, staff concludes that the Atmospheric  
23 Acidification Protection Index form as described in Chapter 5 is best suited to provide for  
24 protection against adverse effects due to acidifying deposition related to  $\text{NO}_x$  and  $\text{SO}_x$ .

25 Within the AAPI, it is also appropriate to consider the specification of values of non-air  
26 quality parameters of the AAPI, including pre-industrial base cation weathering, nitrogen  
27 retention and uptake, runoff, and levels of reduced nitrogen deposition. As discussed in Chapter  
28 5, staff is proposing that the pre-industrial base cation weathering, nitrogen retention and uptake,  
29 and runoff values be determined by assessing the critical loads associated with a specific  
30 percentile of the waterbodies within defined spatial boundaries (as noted in Chapter 5,  
31 consideration is being given to a number of different methods for defining spatial boundaries).

1 The values of pre-industrial base cation weathering, nitrogen retention and uptake, and runoff  
2 values for a selected percentile waterbody are then used as the values in the form of the standard  
3 as realized for the specific ecoregion.

4 The value of reduced nitrogen is initially set using deposition of  $\text{NH}_x$  modeled using the  
5 CMAQ, evaluated for the period 2002-2005. Staff is considering the most appropriate spatial  
6 averaging extent for reduced nitrogen. Figure 9-1 shows spatially interpolated values of reduced  
7 nitrogen deposition based on 12km CMAQ modeling in the Eastern U.S. It is clear that in some  
8 locations, there is significant heterogeneity in reduced nitrogen deposition within ecoregion III  
9 boundaries. Given this information, two possible approaches to estimating reduced nitrogen  
10 values in the AAPI algorithm are 1) average reduced nitrogen deposition within an ecoregion,  
11 acknowledging that this will lead to uncertainties in the level of protection associated with levels  
12 of ambient  $\text{NO}_Y$  and  $\text{SO}_X$ , or 2) allow for additional spatial refinement of sensitive areas to  
13 reflect the heterogeneity of reduced nitrogen deposition. This will result in multiple  
14 parameterizations of the AAPI within a single ecoregion.

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**Figure 9-1. Spatially interpolated CMAQ estimates of deposition of reduced nitrogen (2002-2004 average)**

Unlike other parameters in the AAPI, reduced nitrogen is expected to change significantly over time because of the largely anthropogenic sources of reduced nitrogen deposition. In order to address this potential, we are exploring methods for specifying the standards in a way that would provide for updating the values of reduced nitrogen in the AAPI based on new modeling of reduced nitrogen deposition, following requirements for modeling established by EPA.

#### 1 **9.1.4. Considerations in Defining Options for the AAPI Standard**

2 Conclusions regarding an appropriate range of levels of the AAPI standard will be  
3 informed by considerations of the levels of the target ecological indicator ANC (related to levels  
4 of protection from effects of chronic and episodic acidification), relationships between target  
5 ANC levels and trajectories of recovery over time, and uncertainties in the various elements of  
6 the AAPI that affect the likelihood that the level of protection intended by a particular target  
7 ANC will be realized when atmospheric concentrations of NO<sub>y</sub> and SO<sub>2</sub>+SO<sub>4</sub> fall below the  
8 tradeoff curve for that target ANC. Chapter 5 provides an assessment of the information related  
9 to selection of a target ANC, and the rationale for focusing additional considerations on specific  
10 target ANC levels, including 20 and 50 µeq/L.

11 The “levels” of the ambient indicators are determined by the selection of a level for the  
12 AAPI, as they represent the quantities of the ambient indicators that will result in the specified  
13 level of the AAPI. Those levels will vary for different locations depending on the non-  
14 atmospheric related characteristics of the ecosystem, the level of reduced nitrogen in the  
15 ecosystem, and the atmospheric transformation ratios (T<sub>NO<sub>y</sub></sub> and T<sub>(SO<sub>2</sub>+SO<sub>4</sub>)</sub>).

16 The secondary NAAQS will reflect the public welfare policy judgments of the  
17 Administrator, based on the science, as to the level of air quality which is requisite to protect the  
18 public welfare from any known or anticipated adverse effects associated with the pollutant in the  
19 ambient air. The exposure and risk assessment provide information regarding the effects  
20 associated with a number of different welfare endpoints at different levels of air quality,  
21 expressed in terms of the joint multiyear mean concentrations of NO<sub>y</sub> and SO<sub>2</sub>+SO<sub>4</sub> determined  
22 such that specific levels of ecosystem protection (for example, ANC greater than 50 µeq/L) are  
23 met. Staff also recognizes that in certain naturally acidic ecosystems, even though the ecological  
24 benchmarks are exceeded, e.g. ANC may be quite low; NO<sub>y</sub> and SO<sub>2</sub>+SO<sub>4</sub> are not contributing  
25 to effects because those systems have chronic natural acidity and will not benefit from reductions  
26 in atmospheric deposition. The secondary NAAQS are not intended to provide protection in  
27 these types of naturally acidic systems. As a result, in our determination of appropriate lakes and  
28 streams to include in the populations of critical loads that determine protective NO<sub>y</sub> and  
29 SO<sub>2</sub>+SO<sub>4</sub> levels, we apply filters to remove lakes and streams that are naturally acidic or  
30 acidified due to mine drainage. The secondary NAAQS are focused on providing protection in

1 areas where ambient NO<sub>y</sub> and SO<sub>2</sub>+SO<sub>4</sub> are resulting in effects in ecosystems with low natural  
2 levels of acidification that are highly sensitive to additional inputs of acid deposition.

3 Staff concludes that ecosystem effects of NO<sub>y</sub> and SO<sub>2</sub>+SO<sub>4</sub> deposition in aquatic  
4 ecosystems are an important public welfare effect of concern, based on the types and extent of  
5 ecosystem services likely to be affected by deposition, as well as the location of some aquatic  
6 ecosystems within state and national protected lands, including Class I national parks and  
7 wilderness areas (Chapter 3).

8

#### 9 **9.1.4.1 Target ANC Level**

10 In reaching staff conclusions regarding target ANC levels that are appropriate to consider for an  
11 AAPI-based standard, staff take into account the currently available scientific information  
12 including: evidence from field and laboratory studies, including evidence of effects in highly  
13 sensitive ecosystems and estimates of risk reductions associated with alternative annual standard  
14 levels, as well as the related limitations and uncertainties associated with this information as  
15 presented and discussed more fully in the ISA and RA (US EPA, 2008; US EPA, 2009). In  
16 developing conclusions regarding the target ANC, we evaluated both evidence and risk based  
17 information. In addition, we consider information on target ANC and pH levels used by other  
18 organizations that have established critical loads for protection of aquatic ecosystems from  
19 effects of acidification.

20 We conclude that it is appropriate to define NO<sub>x</sub> and SO<sub>x</sub> standards that will provide  
21 generally consistent protection for acid sensitive lakes and streams across the country. In order  
22 to do so, we focus attention on considering target ANC levels that will lead to a level of AAPI  
23 that provides the same protection for sensitive aquatic ecosystems throughout the U.S. The  
24 result of focusing on a nationally protective level of AAPI is a set of varying NO<sub>y</sub>/(SO<sub>2</sub>+SO<sub>4</sub>)  
25 tradeoff curves across the U.S. reflecting that NO<sub>y</sub> and SO<sub>2</sub>+SO<sub>4</sub> affect acidification in different  
26 ways depending on underlying ecosystem characteristics and levels of reduced nitrogen, such  
27 that the same AAPI is calculated with differing levels of NO<sub>y</sub> and SO<sub>2</sub>+SO<sub>4</sub>. This approach  
28 recognizes that changes in air quality to meet the standards may reflect differing combinations of  
29 NO<sub>y</sub> and SO<sub>2</sub>+SO<sub>4</sub> leading to the same level of the AAPI.

30 Based on our analyses of risks of impacts on aquatic species diversity and fitness and on  
31 the basis of the scientific effects literature, we conclude that achieving a target ANC of 50 µeq/L

1 would substantially decrease the effects of acidification due to NO<sub>y</sub> and SO<sub>2</sub>+SO<sub>4</sub> on aquatic  
2 ecosystems, decreasing the risk of losses in biodiversity and mortality in fish and other aquatic  
3 organisms, and improving the overall health of aquatic ecosystems. Additionally, it is  
4 anticipated that achieving a target ANC of 50 µeq/L would provide increased protection from  
5 NO<sub>y</sub> and SO<sub>2</sub>+SO<sub>4</sub> in areas with higher levels of variability in ecosystem sensitivity due to  
6 variability in meteorology, bedrock geology, topography, land use characteristics, or reduced  
7 nitrogen deposition.

8 It is recognized, however, that a achieving a target ANC of 50 µeq/L would likely not  
9 protect the most sensitive aquatic ecosystems or species within those ecosystems from the effects  
10 of NO<sub>y</sub> and SO<sub>2</sub>+SO<sub>4</sub>. At ANC levels below 100 µeq/L, while overall health of an aquatic  
11 community can be maintained, ANC levels are expected to be such that fish fitness and  
12 community diversity begin to decline. At ANC levels between 100 and 50 µeq/L, ANC levels  
13 are expected to be such that the fitness of sensitive species (e.g., brook trout, zooplankton) also  
14 begins to decline.

#### 15 16 **9.1.4.2 Target Percent of Waterbodies to Meet a Target ANC Level**

17 The appropriate range of levels of AAPI is also informed by the selection of a target  
18 percent of protection for waterbodies within particular acid sensitivity classes or ecoregions (see  
19 Chapter 5). More specifically, the greater percentage of waterbodies that are to be protected  
20 generally indicates a greater likelihood that sensitive waterbodies will achieve a target ANC  
21 level, and as such, in setting the level of the standard, there will be less need to reflect  
22 uncertainty in the likelihood that those sensitive waterbodies will be protected.

23 It is also important to consider that while the target ANC level will not be met in all  
24 sensitive waterbodies if a target percent less than 100 is selected, all waterbodies will realize  
25 some level of protection due to decreases in NO<sub>y</sub> and SO<sub>2</sub>+SO<sub>4</sub> to meet the target ANC in  
26 targeted waterbodies.

27 Additional analyses of the implications of alternative target percentages of waterbodies  
28 are underway and are expected to be completed for inclusion in the final PA.

#### 29 30 **9.1.4.2 Additional Considerations Related to Developing Options for the AAPI Form** 31 **of the Standard**

1 In developing options for the standard, consideration should of the degree to which any  
2 specific AAPI would lead to achieving the desired ANC level, and a judgment as to the degree of  
3 protection of public welfare that is warranted. These considerations should incorporate a wide  
4 number of factors, including the percent of water bodies within acid sensitive areas that the  
5 Administrator determines should be protected at the targeted ANC level, as well as consideration  
6 of achieving desired levels of protection within generational timeframes, e.g., 20 to 40 years,  
7 concerns about protection against episodic acidification events, and uncertainties in the modeling  
8 of critical loads, nitrogen uptake and retention, reduced nitrogen deposition, and relationships of  
9 atmospheric concentrations to deposition. In considering options for the standard that would  
10 reflect consideration for providing requisite welfare protection against know or anticipated  
11 effects, we believe that while the available information is insufficient to set separate standards  
12 for terrestrial acidification and aquatic and terrestrial nutrient enrichment effects, it is also  
13 appropriate to consider the evidence of those effects and the likelihood for co-protection  
14 provided by standards targeted at protection against effects of aquatic acidification (Chapter 6).

15 Chapter 7 provided a summary and synthesis of critical uncertainties and implications for  
16 the standards. While many uncertainties cannot be quantitatively assessed, and as such cannot be  
17 used to recommend specific quantitative changes to the AAPI, there are several uncertainty  
18 analyses which give some insight into the magnitude and direction of the uncertainty. For  
19 example, uncertainty analysis of the MAGIC model of critical loads indicates that modeled pre-  
20 industrial ANC (which informs the distribution of critical loads on which  $\text{NO}_y$  and  $\text{SO}_2+\text{SO}_4$   
21 levels are based by establishing the natural ability of an aquatic ecosystem to neutralize acid  
22 inputs) has 95 percent confidence intervals that are 10 percent higher (or lower) than the mean  
23 estimate for lakes, and 5 percent higher (or lower) than the mean estimate for streams. Similar  
24 uncertainty exists regarding overall uncertainty in the models used to generate critical loads for  
25 determining the  $\text{NO}_y$  and  $\text{SO}_2+\text{SO}_4$  tradeoff curves.

26 The deposition of reduced nitrogen is a critical input to the AAPI and has a large  
27 expected uncertainty due to the use of CMAQ modeling which relies on uncertain chemistry and  
28 uncertain inventories of ammonia emissions. Much of the uncertainty introduced by reduced  
29 nitrogen deposition can be decreased by improvements in measurements of reduced nitrogen  
30 deposition and improvements in the emissions inventories of ammonia and characterization of  
31 ammonia chemistry within the CMAQ modeling. These improvements are underway, and our

1 form of the standard is designed to allow for dynamic updates to reduced nitrogen deposition  
2 parameters.

3  
4

5 **9.1.5 Additional protections for ecosystems against the effects of terrestrial acidification**  
6 **and terrestrial and aquatic nutrient enrichment**

7

8 While we are not basing the elements of this standard primarily on consideration of  
9 effects other than aquatic acidification, our approach recognizes that some level of protection  
10 against effects of acidification in terrestrial ecosystems and effects of nutrient enrichment in  
11 terrestrial and aquatic ecosystems is likely to be realized through changes in air quality to meet  
12 the AAPI standard. We recognize that an annual standard focused on aquatic acidification  
13 cannot be expected to offer protection against all of the welfare effects from NO<sub>y</sub> and SO<sub>2</sub>+SO<sub>4</sub>,  
14 especially in areas that are sensitive to nutrient enrichment but are not acid sensitive. However,  
15 based on the information available in the ISA and REA, we conclude that the available  
16 information is not sufficient to set a complementary standard to provide protection against  
17 additional effects of NO<sub>y</sub> through nutrient enrichment, and that additional research is necessary  
18 to support the setting of such a standard, especially in the areas of identifying the specific  
19 impacts of decreases in atmospheric nitrogen deposition (see chapter 7). CASAC has noted that  
20 our current framework, with appropriate modifications, should be applicable to developing  
21 standards to provide protection against acidification effects in sensitive terrestrial ecosystems.  
22 However, staff has concluded that the current data is not sufficient to develop a separate  
23 terrestrial acidification based standard at this time. A primary limitation is the identification of  
24 specific levels of harm associated with the BC/Al ecological indicator.

25 Sensitive terrestrial ecosystems that are located in watersheds with acid sensitive water  
26 bodies are likely to receive the most protection under an aquatic acidification targeted standard.  
27 Terrestrial ecosystems outside of these watersheds are likely to see some level of protection, but  
28 will not realize targeted changes in ambient NO<sub>y</sub> and SO<sub>2</sub>+SO<sub>4</sub> (Chapter 6).

29 Chapter 6 evaluated the relative protection for terrestrial ecosystems in areas from  
30 meeting a target ANC of 50 µeq/L compared to meeting a target Bc:Al ratio of 10. Critical  
31 loads for N and S were compared to determine which of the targets would result in a more

1 stringent critical load. Over half of the watersheds had a lower critical load to meet the ANC  
2 target compared to the critical load to meet the Bc:Al ratio target. As a result, those watersheds  
3 are likely to be protected from both terrestrial and aquatic impacts when the ANC target is met.  
4 When the water bodies are more sensitive to deposition (“highly sensitive” or “moderately  
5 sensitive”), the aquatic critical acid loads generally provide a greater level of protection against  
6 acidifying nitrogen and sulfur deposition in the watershed.

7 The tradeoff curves for NO<sub>y</sub> and SO<sub>2</sub>+SO<sub>4</sub> that are associated with protection against  
8 aquatic acidification also provide bounding conditions for nitrogen that can be compared against  
9 benchmarks of effects associated with nitrogen deposition in sensitive terrestrial and aquatic  
10 ecosystems. Achieving an ANC level of 50 µeq/L for 90 percent of lakes and streams  
11 nationwide would provide some protection against leaching in northeast forests, but would need  
12 to be lower to protect California coastal sage scrub, lichens in mixed conifer forests, alpine lake  
13 communities, and Minnesota grasslands (Section 6.2). In the case of aquatic nutrient  
14 enrichment, comparison of maximum allowable NO<sub>x</sub> levels on the tradeoff curves with  
15 deposition requirements to meet the Chesapeake Bay TMDL shows that without further  
16 restrictions on NO<sub>y</sub> concentrations, standards set to protect against aquatic acidification will not  
17 be protective against effects of aquatic nutrient enrichment in the Chesapeake Bay.

### 18

### 19 **9.1.6 Summary of options**

### 20

21 To facilitate evaluation of the elements of the standard and staff conclusions regarding  
22 those elements, we have constructed a summary table showing the elements of the AAPI, options  
23 for each element, and staff conclusions where appropriate.

24

25 [Table 9-1 to be provided]

26

## 27 **9.2 CONCLUSIONS**

28 The following secondary NAAQS conclusions encompass the breadth of policy-relevant  
29 considerations described in this policy assessment. We note that staff conclusions to be  
30 presented in the final PA will consider input received from CASAC and the public on this second  
31 draft PA. We recognize that selecting from among alternative standards will necessarily reflect

1 consideration of the qualitative and quantitative uncertainties inherent in the relevant evidence  
2 and in the assumptions that underlie the quantitative risk assessment. In identifying these  
3 alternative secondary standards and ranges of levels for consideration, we are mindful that the  
4 Clean Air Act requires standards to be set that are requisite to protect public from known or  
5 anticipated adverse effects, such that the standards are to be neither more nor less stringent than  
6 necessary. Thus, the Act does not require that the NAAQS be set at no effect levels, but rather at  
7 levels that avoid adverse effects on public welfare:

8 (1) Based on the policy-relevant findings from the ISA described in Chapter 2, and while  
9 recognizing that important uncertainties and research questions remain, staff conclude that great  
10 progress has been made since the last reviews of the secondary standards for NO<sub>x</sub> and SO<sub>x</sub>. We  
11 generally find support in the available effects-based evidence for consideration of NO<sub>x</sub> and SO<sub>x</sub>  
12 standards that are at least as protective as the current standard and do not find support for  
13 consideration of NO<sub>x</sub> and SO<sub>x</sub> standards that are less protective than the current standard. The  
14 staff also concludes that consideration of joint standards for NO<sub>x</sub> and SO<sub>x</sub> is appropriate given  
15 the common atmospheric processes governing the deposition of NO<sub>x</sub> and SO<sub>x</sub> to sensitive  
16 ecosystems, and given the combined effects of N and S deposition on acidification of soil and  
17 water.

18 (2) Staff concludes that ambient NO<sub>x</sub> is a significant component of atmospheric nitrogen  
19 deposition, even in areas with relatively high rates of deposition of reduced nitrogen. Staff make  
20 this conclusion based on the analysis in Chapter 3 of the REA, which provides a thorough  
21 assessment of the contribution of NO<sub>x</sub> to nitrogen deposition throughout the U.S., and the  
22 relative contributions of ambient NO<sub>x</sub> and reduced forms of nitrogen.

23 (3) Staff concludes based on the case study results provided in the REA, that current  
24 levels of NO<sub>x</sub> and SO<sub>x</sub> are associated with deposition that leads to ANC values below  
25 benchmark values that cause ecological harm and losses in ecosystem services. Staff concludes  
26 that the evidence and risk assessment support strongly a relationship between atmospheric  
27 deposition of NO<sub>x</sub> and SO<sub>x</sub> and ANC, and that ANC is an excellent indicator of aquatic  
28 acidification. Staff also concludes that at levels of deposition associated with NO<sub>x</sub> and SO<sub>x</sub>  
29 concentrations at or below the current standards, ANC levels are expected to be below  
30 benchmark values that are associated with significant losses in fish species richness, which is  
31 associated with reductions in recreational fishing services. Although there are many other

1 ecosystem services potentially affected by reductions in ANC, including subsistence fishing,  
2 natural habitat provision, and biological control, confidence in the specific translation of ANC  
3 values to these additional ecosystem services is much lower.

4 (4) Losses in aquatic resources associated with ANC levels below 50  $\mu\text{eq/L}$  are clearly  
5 associated with significant losses in economic value. Based on the best available data, just in  
6 New York., increasing ANC levels to 50 in the Adirondacks is estimated to result in \$300 to  
7 \$800 million in annual benefits in 2006 dollars. This estimate represents only a fraction of the  
8 total economic value of ecosystem damages as many impacted resources are not amenable to  
9 economic valuation methods. In addition, economic damages are also likely to occur in other  
10 areas affected by acidification, including New England, the Appalachian Mountains (northern  
11 Appalachian Plateau and Ridge/Blue Ridge region), and the Upper Midwest. Staff concludes  
12 that reducing acidifying deposition of  $\text{NO}_x$  and  $\text{SO}_x$  will result in improvements in public  
13 welfare by increasing the quantity and quality of ecosystem services, including recreational  
14 fishing and other services associated with improved water quality.

15 (5) Staff initially concludes based on the case study results that current levels of ambient  
16  $\text{NO}_x$  and  $\text{SO}_x$  are associated with deposition that leads to Bc:Al values below benchmark values  
17 that cause ecological harm and losses in ecosystem services. Staff concludes that the evidence  
18 and risk assessment support strongly a relationship between atmospheric deposition of  $\text{NO}_x$  and  
19  $\text{SO}_x$  and Bc:Al, and that Bc:Al is a good indicator of terrestrial acidification. Staff also  
20 concludes that at levels of deposition associated with  $\text{NO}_x$  and  $\text{SO}_x$  concentrations at or below  
21 the current standards, Bc:Al levels are expected to be below benchmark values that are  
22 associated with significant losses in tree health and growth, which are associated with reductions  
23 in timber production. While there are many other ecosystem services, including maple syrup  
24 production, natural habitat provision, and regulation of water, climate, and erosion, potentially  
25 affected by reductions in Bc:Al, confidence in the specific translation of Bc:Al values to these  
26 additional ecosystem services is much lower.

27 (6) On the basis of the acidification and nutrient enrichment effects that have been  
28 observed to still occur under current ambient conditions and those predicted to occur under the  
29 scenario of just meeting the current secondary NAAQS, staff concludes that the current  
30 secondary NAAQS are inadequate to protect the public welfare from known and anticipated  
31 adverse welfare effects from aquatic and terrestrial acidification associated with deposition of

1 NO<sub>x</sub> and SO<sub>x</sub>. As discussed above, this conclusion derives from several lines of evidence. Staff  
2 also concludes that the current NO<sub>x</sub> and SO<sub>x</sub> secondary standards are adequate to protect against  
3 direct gas-phase effects on vegetation, and as such, should be retained to preserve protection  
4 against these welfare effects.

5 (7) Staff has concluded, based on the completeness of the available evidence and  
6 quantitative risk information, that effects due to aquatic acidification are most suitable for  
7 defining additional secondary standards for NO<sub>x</sub> and SO<sub>x</sub>. Staff notes that in developing a  
8 standard designed to protect against the effects of aquatic acidification due to deposition of NO<sub>x</sub>  
9 and SO<sub>x</sub>, the resulting standards may not provide protection against known effects associated  
10 with terrestrial acidification and with nutrient enrichment in sensitive aquatic and terrestrial  
11 ecosystems.

12 (8) It is appropriate to consider using indicators other than NO<sub>2</sub> and SO<sub>2</sub> as the indicators  
13 for an additional standard that is intended to address the ecological effects associated with  
14 deposition of NO<sub>x</sub> and SO<sub>x</sub> to sensitive ecosystems. Given the reasons discussed in Chapters 2,  
15 4, and 5 of this policy assessment, staff concludes that NO<sub>x</sub> (oxides of nitrogen, the definition in  
16 section 302 (v) of the CAA), is best represented by the atmospheric indicator NO<sub>y</sub>, defined as  
17 NO<sub>2</sub> + NO + HNO<sub>3</sub> + PAN + 2N<sub>2</sub>O<sub>5</sub> + HONO + NO<sub>3</sub> + organic nitrates + particulate NO<sub>3</sub> is the  
18 more appropriate indicator of oxides of nitrogen, and that SO<sub>2</sub>+SO<sub>4</sub> is the more appropriate  
19 indicator of oxides of sulfur for purposes of a secondary standard addressing aquatic  
20 acidification.

21 (9) It is appropriate to use multi-year averages of concentrations of NO<sub>y</sub> and SO<sub>2</sub>+SO<sub>4</sub> as  
22 the averaging times for the secondary standards, based on the chronic nature of acidification, and  
23 the protection against episodic acidification provided by a standard based on annual average  
24 concentrations. Averaging periods in the range of 3 to 5 years are most appropriate.

25 (10) It is appropriate to consider adding a new standard with a different form for NO<sub>y</sub>  
26 and SO<sub>2</sub>+SO<sub>4</sub> as the current form does not take into account the linkages between NO<sub>x</sub> and SO<sub>x</sub>  
27 in the causation of effects associated with acidification of aquatic ecosystems. Based on the  
28 causal linkages between NO<sub>x</sub> and SO<sub>x</sub>, deposition of N and S, and the indicator of acidification,  
29 ANC, staff concludes that a standard with a form specified as an atmospheric acidification  
30 protection index (AAPI) should be added. A standard with this form would reflect the important

1 roles of underlying ecosystem characteristics, determinants of deposition, and deposition of  
2 reduced nitrogen in determining the potential effects from deposition of NO<sub>x</sub> and SO<sub>x</sub>.

3 (11) Staff has concluded, based on the evidence and risk based information, and  
4 consideration of information related to definitions of adversity, that

5 a) a target level of ANC of 20 µeq/L will protect against significant losses in  
6 fish mortality in many sensitive lakes, but will place less weight on protection against  
7 losses in aquatic biodiversity, and will be less protective against potential acidification  
8 episodes,

9 b) a target level of ANC of 50 µeq/L will protect against significant mortality  
10 in aquatic organisms and loss of fish health and biodiversity in sensitive lakes and  
11 streams, and will give weight to considerations of uncertainties in the time to recovery of  
12 aquatic ecosystems,

13 c) target levels of ANC above 50 µeq/L may provide additional protection  
14 against declines in fitness of sensitive species (e.g., brook trout, zooplankton), however,  
15 overall health of aquatic communities may not be impacted.

16

1 **9.3 REFERENCES**

2

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14

**APPENDIX A**  
**ADDITIONAL INFORMATION FOR CHAPTER 5**

**A.1 Conceptual Design of the Standard**

This is supplemental information to support the discussion of the conceptual design of the standard that is presented in Chapter 5 of the Policy Assessment Document. The aquatic acidification analyses developed in the REA used a number of different models and calculation techniques that are important for the development of the standard. The goal of this Appendix is to summarize information from the REA analysis that is most relevant to the Policy Assessment. A brief summary of the REA analyses are presented in section A.1.1. In section A.1.2 there is a general summary and technical discussion of the critical loads modeling approaches that were used in the REA, followed by a brief description of MAGIC model data requirements.

**A.1.1 Technical summary of methods used in the REA Aquatic Acidification analysis**

The aquatic acidification analysis is presented in Chapter 4 and Appendix 4 of the REA. The analysis uses multiple techniques to show the relationship between ANC and NOx and SOx deposition, as well as determine the current level of risk to water bodies that occur in sensitive areas. A brief summary of the techniques and objectives of the REA analysis is given in Table A-1.

<b>Table A-1. Brief summary of objects and methods used in the REA Aquatic Acidification analysis.</b>		
<b>Technique</b>	<b>Objectives</b>	
Time-series graphs of current conditions	1	Data from monitoring networks collected from 1990 to 2006 were plotted to show trends in concentrations of pollutants, deposition and acidification for each case study site. The data included surface water concentration of nitrate, sulfate and ANC; deposition of sulfate and nitrate; as well as air concentration of SOx, NOx and NH4
MAGIC	1	Used to estimate the relationship between ANC values and anthropogenic NOx and SOx emission from the past (preacidification ~1860), present (2002 and 2006) and projected into the future (2020 and 2050). Analysis included 44 lakes from Adirondacks and 60 streams from Shenandoah.
	2	Used to develop input parameters for critical loads modeling (i.e. weathering rates)
	3	Used for uncertainty analysis
Critical Loads modeling	1	SSWC and FAB models used to calculate critical loads for critical limits of ANC = 0, 20, 50, 100

<b>Table A-1. Brief summary of objects and methods used in the REA Aquatic Acidification analysis.</b>		
	2	Critical loads for ANC critical limits calculated for 169 lakes in the Adirondacks and 60 streams in the Shenandoah using water quality data from monitoring sites collected in 2006
	3	Critical loads exceedences calculated by comparing the critical loads that were calculated by SSWC with deposition data from NADP for wet deposition and CMAQ for dry deposition, both for the year 2002
Regional Extrapolation	1	117 of the critical loads calculated for the Adirondacks were extrapolated to lakes defined by the New England EMAP probability survey, representing 1842 lakes, to infer the # of lakes that exceeded their critical load
	2	69 of the critical loads calculated for the Shenandoah were extrapolated to 330 streams based on bed rock geology classification.

1

2 **A.1.2 Technical summary of critical loads modeling in the REA**

3 The critical load of acidity for lakes or streams was derived from present-day  
 4 water chemistry using a combination of steady-state models. Both the Steady-State Water  
 5 Chemistry (SSWC) model and First-order Acidity Balance model (FAB) is based on the  
 6 principle that excess base-cation production within a catchment area should be equal to or  
 7 greater than the acid anion input, thereby maintaining the ANC above a preselected level  
 8 (Reynolds and Norris, 2001; Posch et al. 1997). These models assume steady-state  
 9 conditions and assume that all  $\text{SO}_4^{2-}$  in runoff originates from sea salt spray and  
 10 anthropogenic deposition. Given a critical ANC protection level, the critical load of  
 11 acidity is simply the input flux of acid anions from atmospheric deposition (i.e., natural  
 12 and anthropogenic) subtracted from the natural (i.e., preindustrial) inputs of base cations  
 13 in the surface water. Final Risk and Exposure Assessment September 2009 Appendix 4,  
 14 Attachment A – 15 *Aquatic Acidification Case Study* Atmospheric deposition of  $\text{NO}_x$  and  
 15  $\text{SO}_x$  contributes to acidification in aquatic ecosystems through the input of acid anions,  
 16 such as  $\text{NO}_3^-$  and  $\text{SO}_4^{2-}$ . The acid balance of headwater lakes and streams is controlled by  
 17 the level of this acidifying deposition of  $\text{NO}_3^-$  and  $\text{SO}_4^{2-}$  and a series of biogeochemical  
 18 processes that produce and consume acidity in watersheds. The biotic integrity of  
 19 freshwater ecosystems is then a function of the acid-base balance, and the resulting  
 20 acidity-related stress on the biota that occupy the water. The calculated ANC of the  
 21 surface waters is a measure of the acid-base balance:

22

23 
$$\text{ANC} = [\text{BC}]^* - [\text{AN}]^* \quad (1)$$

24

1 where [BC]\* and [AN]\* are the sum of base cations and acid anions (NO<sub>3</sub><sup>-</sup> and SO<sub>4</sub><sup>2-</sup>),  
2 respectively. Equation (1) forms the basis of the linkage between deposition and surface  
3 water acidic condition and the modeling approach used. Given some “target” ANC  
4 concentration [ANClimit]) that protects biological integrity, the amount of deposition of  
5 acid anions (AN) or depositional load of acidity CL(A) is simply the input flux of acid  
6 anions from atmospheric deposition that result in a surface water ANC concentration  
7 equal to the [ANClimit] when balanced by the sustainable flux of base cations input and  
8 the sinks of nitrogen and sulfur in the lake and watershed catchment.

9  
10 Critical loads for nitrogen and sulfur (CL(N) + CL(S) ) or critical load of acidity  
11 CL(A) were calculated for each waterbody from the principle that the acid load should  
12 not exceed the nonmarine, nonanthropogenic base cation input and sources and sinks in  
13 the catchment minus a neutralizing to protect selected biota from being damaged:

14  
15 
$$CL(N) + CL(S) \text{ or } CL(A) = BC^*_{dep} + BC_w - B_{cu} - AN - ANClimit \text{ (2)}$$

16  
17 Where,

18  $BC^*_{dep} = (BC^* = Ca^* + Mg^* + K^* + Na^*)$ , nonanthropogenic deposition flux of base cations

19  $BC_w$  = the average weathering flux, producing base cations

20  $B_{cu}$  ( $B_c = Ca^* + Mg^* + K^*$ ) = the net long-term average uptake flux of base cations in the  
21 biomass (i.e., the annual average removal of base cations due to harvesting)

22  $AN$  = the net long-term average uptake, denitrification, and immobilization of nitrogen  
23 anions (e.g. NO<sub>3</sub><sup>-</sup>) and uptake of SO<sub>4</sub><sup>2-</sup>

24  $ANClimit$  = the lowest ANC-flux that protects the biological communities.

25  
26 Since the average flux of base cations weathered in a catchment and reaching the  
27 lake or streams is difficult to measure or compute from available information, the average  
28 flux of base cations and the resulting critical load estimation were derived from water  
29 quality data (Henriksen and Posch, 2001; Henriksen et al., 1992; Sverdrup et al., 1990).  
30 Weighted annual mean water chemistry values were used to estimate average base cation  
31 fluxes, which were calculated from water chemistry data collected from the Temporally  
32 Integrated Monitoring of Ecosystems (TIME)/Long-Term Monitoring (LTM) monitoring

1 networks, that include Adirondack Longterm Monitoring (ALTM), Virginia Trout Stream  
2 Sensitivity Study (VTSSS), and the Shenandoah Watershed Study (SWAS), and  
3 Environmental Monitoring and Assessment Program (EMAP) (see Section 4.1.2.1 of  
4 Chapter 4).

5  
6 The preacidification nonmarine flux of base cations for each lake or stream,  $BC^*0$ , is

7  
8  $BC^*0 = BC^*dep + BC_w - B_{cu}$  (3)  
9

10 Thus, critical load for acidity can be rewritten as

11  
12  $CL(N) + CL(S) = BC^*0 - AN - AN_{limit} = Q \cdot ([BC^*]_0 - [AN] - [ANC]_{limit})$ , (4)  
13

14 where the second identity expresses the critical load for acidity in terms of catchment  
15 runoff ( $Q$ ) m/yr and concentration ( $[x] = X/Q$ ). The sink of nitrogen in the watershed is  
16 equal to the uptake ( $N_{upt}$ ), immobilization ( $N_{imm}$ ), and denitrification ( $N_{den}$ ) of  
17 nitrogen in the catchment. Thus, critical load for acidity can be rewritten as

18  
19  $CL(N) + CL(S) = \{fN_{upt} + (1 - r)(N_{imm} + N_{den})\} + ([BC^*]_0 - [ANC]_{limit})Q$  (5)  
20

21 where  $f$  and  $r$  are dimensionless parameters that define the fraction of forest cover in the  
22 catchment and the lake/catchment ratio. The in-lake retention of nitrogen and sulfur was  
23 assumed to be negligible. Equation 5 described the FAB model that was applied when  
24 sufficient data was available to estimate the uptake, immobilization, and denitrification of  
25 nitrogen and the neutralization of acid anions (e.g.  $NO_3^-$ ) in the catchment. In the case  
26 where data was not available, the contribution of nitrogen anions to acidification was  
27 assumed to be equal to the nitrogen leaching rate ( $N_{leach}$ ) into the surface water. The  
28 flux of acid anions in the surface water is assumed to represent the amount of nitrogen  
29 that is not retained by the catchment, which is determined from the sum of measured  
30 concentration of  $NO_3^-$  and ammonia in the stream chemistry. This case describes the  
31 SSWC model and the critical load for acidity is

32  
33  $CL(A) = Q \cdot ([BC^*]_0 - [ANC]_{limit})$  (6)  
34

35 where the contribution of acid anions is considered as part of the exceedances calculation  
36 (see Section 1.2.5, below). For the assessment of current condition in both case study

1 areas, the critical load calculation described in Equation 6 was used for most lakes and  
2 streams. The lack of sufficient data for quantifying nitrogen denitrification and  
3 immobilization prohibited the wide use of the FAB model. In addition, given the  
4 uncertainty in quantifying nitrogen denitrification and immobilization, the flux of  
5 nitrogen anions in the surface water was assumed to more accurately reflect the  
6 contribution of NO<sub>3</sub><sup>-</sup> to acidification. Several major assumptions are made: (1) steady-  
7 state conditions exist, (2) the effect of nutrient cycling between plants and soil is  
8 negligible, (3) there are no significant nitrogen inputs from sources other than  
9 atmospheric deposition, (4) ammonium leaching is negligible because any inputs are  
10 either taken up by biota or adsorbed onto soils or nitrate compounds, and (5) longterm  
11 sinks of sulfate in the catchment soils are negligible.

12

#### 13 ***A.1.2.1 Preindustrial Base Cation Concentration***

14 Present-day surface water concentrations of base cations are elevated above their  
15 steady-state preindustrial concentrations because of base cation leaching through ion  
16 exchange in the soil due to anthropogenic inputs of SO<sub>4</sub><sup>2-</sup> to the watershed. For this  
17 reason, present-day surface water base cation concentrations are higher than natural or  
18 preindustrial levels, which, if not corrected for, would result in critical load values not in  
19 steady-state condition. To estimate the preacidification flux of base cations, the present  
20 flux of base cations was estimated,

21

22 BC\*<sub>t</sub>, given by  $BC^*_{t} = BC^*_{dep} + BC_{w} - B_{cu} + BC_{exc}$ , (7)

23

24 Where BC<sub>exc</sub> = the release of base cations due to ion-exchange processes. Assuming  
25 that deposition, weathering rate, and net uptake have not changed over time, BC<sub>exc</sub> can  
26 be obtained by subtracting Equation 5 from Equation 7:

27

28  $BC_{exc} = BC^*_{t} - BC^*_{0}$  (8)

29

30 This present-day excess production of base cations in the catchment was related to the  
31 long-term changes in inputs of nonmarine acid anions ( $\Delta SO^*_{2} + \Delta NO_{3}$ ) by the F-factor  
32 (see below):

1

2  $BC_{exc} = F (\Delta SO_4^{*2} + \Delta NO_3)$  (9)

3

4 For the preacidification base cation flux, solving Equation 5 for  $BC^*0$  and then  
5 substituting Equation 8 for  $BC_{exc}$  and explicitly describing the long-term changes in  
6 nonmarine acid ion inputs:

7

8  $BC^*0 = BC^*t - F (SO_4^{*4,t} - SO_4^{*4,0} + NO_3^{*3,t} - NO_3^{*3,0})$  (10)

9

10 The preacidification  $NO_3^-$  concentration,  $NO_3^{*3,0}$ , was assumed to be zero.

11

#### 12 **A.1.2.2 F-factor**

13 An F-factor was used to correct the concentrations and estimate preindustrial base  
14 concentrations for lakes in the Adirondack Case Study Area. In the case of streams in the  
15 Shenandoah Case Study Area, the preindustrial base concentrations were derived from  
16 the MAGIC model as the base cation supply in 1860 (hindcast) because the F-factor  
17 approach is untested in this region. An F-factor is a ratio of the change in nonmarine base  
18 cation concentration due to changes in strong acid anion concentrations (Henriksen,  
19 1984; Brakke et al., 1990):

20

21  $F = ([BC^*]_t - [BC^*]_0) / ([SO_4^{*4}]_t - [SO_4^{*4}]_0 + [NO_3^{*3}]_t - [NO_3^{*3}]_0)$ , (12)

22

23 where the subscripts t and 0 refer to present and preacidification conditions, respectively.  
24 If  $F=1$ , all incoming protons are neutralized in the catchment (only soil acidification); at  
25  $F=0$ , none of the incoming protons are neutralized in the catchment (only water  
26 acidification). The F-factor was estimated empirically to be in the range 0.2 to 0.4, based  
27 on the analysis of historical data from Norway, Sweden, the United States, and Canada  
28 (Henriksen, 1984). Brakke et al. (1990) later suggested that the F-factor should be a  
29 function of the base cation concentration:

30

31  $F = \sin (\pi/2 Q[BC^*]_t/[S])$  (13)

1

2 where

3  $Q$  = the annual runoff (m/yr).  $[S]$  = the base cation concentration at which  $F=1$ ; and for  
4  $[BC^*]_t > [S]$   $F$  is set to 1. For Norway  $[S]$  has been set to 400 milliequivalents per cubic  
5 meter (meq/m<sup>3</sup>)(circa.8 mg Ca/L) (Brakke et al., 1990). The preacidification SO<sub>4</sub><sup>2-</sup>  
6 concentration in lakes,  $[SO_4^*]_0$ , is assumed to consist of a constant atmospheric  
7 contribution and a geologic contribution proportional to the concentration of base cations  
8 (Brakke et al., 1989). The preacidification SO<sub>4</sub><sup>2-</sup> concentration in lakes,  $[SO_4^*]_0$  was  
9 estimated from the relationship between  $[SO_4^*]_0$  and  $[BC]_t^*$  based on work completed  
10 by Henriksen et al., 2002 as described by the following equation:

11

12  $[SO_4^*]_0 = 15 + 0.16 * [BC]_t^*$  (14)

13

14

1  
2

<b>Table A-2 Illustrates SSWC Approach – Environmental Variables</b>			
$CL(A) = BC_{dep}^* + BC_w - BC_u - ANC_{limit}$ $CL(A) = Q \cdot ([BC^*]_0 - [ANC]_{limit})$			
	<b>Variable Code</b>	<b>Description</b>	<b>Source</b>
1	$BC_{dep}^*$	Sum (Ca*+Mg*+K*+Na*), nonanthropogenic deposition flux of base cations	Wet NADP and Dry CASTNET
2	$BC_w$	Average weathering flux of base cations	Calculated (5-17)
3	$Bc_u$	Sum (Ca+Mg+K), the net long-term average uptake flux of base cations in the biomass	USFS-FIA data
4	$ANC_{limit}$	Lowest ANC-flux that protects the biological communities	Set
5	$Ca^*$	Sea Salt corrected Surface water concentration ( $\mu\text{eq/L}$ ) growing season average. ( $Ca - (CL \times 0.0213)$ )	Water quality data
6	$Mg^*$	Sea Salt corrected Surface water concentration ( $\mu\text{eq/L}$ ) growing season average. ( $Mg - (CL \times 0.0669)$ )	Water quality data
7	$Na^*$	Sea Salt corrected Surface water concentration ( $\mu\text{eq/L}$ ) growing season average. ( $Na - (CL \times 0.557)$ )	Water quality data
8	$K^*$	Sea Salt corrected Surface water concentration ( $\mu\text{eq/L}$ ) growing season average. ( $K - (CL \times 0.0206)$ )	Water quality data
9	$SO_4^*$	Sea Salt corrected Surface water concentration ( $\mu\text{eq/L}$ ) growing season average. ( $SO_4 - (CL \times 0.14)$ )	Water quality data
10	CL	Surface water concentration ( $\mu\text{eq/L}$ ) growing season average.	Water quality data
11	$SO_4^*$	Surface water concentration ( $\mu\text{eq/L}$ ) growing season average.	Water quality data
12	$NO_3^*$	Surface water concentration ( $\mu\text{eq/L}$ ) growing season average.	Water quality data
13	Q	The annual runoff (m/yr)	USGS
14	$[BC^*]_0$	Preindustrial flux of base cations in surface water, corrected for sea salts	Calculated from water quality data
15	$[SO_4^*]_0$	Preindustrial flux of sulfate in surface water, corrected for sea salts	Estimated
16	$[NO_3^*]_0$	Preindustrial flux of nitrate, corrected for sea salts	Equal to 0
17	F	Calculated factor	Fix values

3

<b>Table A-2 FAB Approach – Environmental Variables</b>			
$DL(N) + DL(S) = \{fN_{upt} + (1 - r)(N_{imm} + N_{den}) + (N_{ret} + S_{ret})\} + ([BC]_0^* - [ANC_{limit}])Q$			
	<b>Variable Code</b>	<b>Description</b>	<b>Source</b>
1	Ndepo	Total N deposition	NADP/CMAQ
2	ANCLimit	Lowest ANC-flux that protects the biological communities	Set
3	[BC*] <sub>0</sub>	Preindustrial flux of base cations in surface water, corrected for sea salt	Calculated from water quality data
4	Ca*	Sea Salt corrected Surface water concentration (µeq/L) growing season average. (Ca – (CL × 0.0213))	Water quality data
5	Mg*	Sea Salt corrected Surface water concentration (µeq/L) growing season average. (Mg – (CL × 0.0669))	Water quality data
6	Na*	Sea Salt corrected Surface water concentration (µeq/L) growing season average. (Na – (CL × 0.557))	Water quality data
7	K*	Sea Salt corrected Surface water concentration (µeq/L) growing season average. (K – (CL × 0.0206))	Water quality data
8	SO <sub>4</sub> *	Sea Salt corrected Surface water concentration (µeq/L) growing season average. (SO <sub>4</sub> – (CL × 0.14))	Water quality data
9	CL	Surface water concentration (µeq/L) growing season average.	Water quality data
10	SO <sub>4</sub> *	Surface water concentration (µeq/L) growing season average.	Water quality data
11	NO <sub>3</sub> *	Surface water concentration (µeq/L) growing season average.	Water quality data
12	Q	The annual runoff (m/yr)	USGS
13	f	f is a dimensionless parameter that define the fraction of forest cover in the catchment	
14	r	r is a dimensionless parameter that define the lake/catchment ratio	
14	N <sub>ret</sub>	The in-lake retention of nitrogen	Estimated
15	S <sub>ret</sub>	The in-lake retention of sulfur	Estimated
16	N <sub>upt</sub>	The net long-term average uptake flux of N in the biomass	USFS-FIA data
17	N <sub>imm</sub>	Immobilization of N in the soils	Estimated fix value
18	N <sub>den</sub>	Denitrification	Estimated fix value
19	Lake Size	Lake size (ha)	DLMs
20	WSH	Watershed area (ha)	Calculated

1  
2

1  
2 **Data requirements for MAGIC**

3         The MAGIC model (Cosby et al., 1985a; 1985b; 1985c) is a mathematical model  
4 (a lumped-parameter model) of soil and surface water acidification in response to  
5 atmospheric deposition based on process-level information about acidification. A process  
6 model, such as MAGIC, characterizes acidification into (1) a section in which the  
7 concentrations of major ions are assumed to be governed by simultaneous reactions  
8 involving  $\text{SO}_4^{2-}$  adsorption, cation exchange, dissolution-precipitation- speciation of  
9 aluminum, and dissolution-speciation of inorganic carbon; and (2) a mass balance section  
10 in which the flux of major ions to and from the soil is assumed to be controlled by  
11 atmospheric inputs, chemical weathering, net uptake and loss in biomass and losses to  
12 runoff. At the heart of MAGIC is the size of the pool of exchangeable base cations in the  
13 soil. As the fluxes to and from this pool change over time owing to changes in  
14 atmospheric deposition, the chemical equilibria between soil and soil solution shift to  
15 give changes in surface water chemistry. The degree and rate of change of surface water  
16 acidity thus depend both on flux factors and the inherent characteristics of the affected  
17 soils.

18         There are numerous input data required to run MAGIC making it rather data  
19 intensive. Atmospheric deposition fluxes for the base cations and strong acid anions are  
20 required as inputs to the model. These inputs are generally assumed to be uniform over  
21 the catchment. The volume discharge for the catchment must also be provided to the  
22 model. In general, the model is implemented using average hydrologic conditions and  
23 meteorological conditions in annual simulations, i.e., mean annual deposition,  
24 precipitation and lake discharge are used to drive the model. Values for soil and surface  
25 water temperature, partial pressure of carbon dioxide and organic acid concentrations  
26 must also be provided at the appropriate temporal resolution.

27         The aggregated nature of the model requires that it be calibrated to observed data  
28 from a system before it can be used to examine potential system response. Calibrations  
29 are based on volume weighted mean annual or seasonal fluxes for a given period of  
30 observation. The length of the period of observation used for calibration is not arbitrary.  
31 Model output will be more reliable if the annual flux estimates used in calibration are

1 based on a number of years rather than just one year. There is a lot of year-to-year  
2 variability in atmospheric deposition and catchment runoff. Averaging over a number of  
3 years reduces the likelihood that an “outlier” year (very dry, etc.) is used to specify the  
4 primary data on which model forecasts are based. On the other hand, averaging over too  
5 long a period may remove important trends in the data that need to be simulated by the  
6 model.

7 The calibration procedure requires that stream water quality, soil chemical and  
8 physical characteristics, and atmospheric deposition data be available for each catchment.  
9 The water quality data needed for calibration are the concentrations of the individual base  
10 cations (Ca, Mg, Na, and K) and acid anions (Cl,  $\text{SO}_4^{2-}$ , and  $\text{NO}_3^-$ ) and the pH. The soil  
11 data used in the model include soil depth and bulk density, soil pH, soil cation-exchange  
12 capacity, and exchangeable bases in the soil (Ca, Mg, Na, and K). The atmospheric  
13 deposition inputs to the model must be estimates of total deposition, not just wet  
14 deposition. In some instances, direct measurements of either atmospheric deposition or  
15 soil properties may not be available for a given site with stream water data. In these  
16 cases, the required data can often be estimated by: (a) assigning soil properties based on  
17 some landscape classification of the catchment; and (b) assigning deposition using model  
18 extrapolations from some national or regional atmospheric deposition monitoring  
19 network. Soil data for model calibration are usually derived as aeri ally averaged values  
20 of soil parameters within a catchment. If soils data for a given location are vertically  
21 stratified, the soils data for the individual soil horizons at that sampling site can be  
22 aggregated based on horizon, depth, and bulk density to obtain single vertically  
23 aggregated values for the site, or the stratified data can be used directly in the model.

### 24 25 26 **A.1.3 Example of the two ways to calculate $N_{\text{ECO}}$ from the first draft $\text{NO}_x$ and $\text{SO}_x$** 27 **secondary NAAQS Policy Assessment Document** 28

29 The steady-state critical load model suggest for use in the NAAQS by the PA  
30 could be constrained by a quantity of N which would be taken up, immobilized or  
31 denitrified by ecosystems and used to adjust the quantity of deposition required to meet a  
32 specified critical load. This term is abbreviated by  $N_{\text{eco}}$ , and could be derived multiple

1 ways. The first is by taking the mean value calculated to represent the long-term amount  
2 of N an ecosystem can immobilize and denitrify before leaching (i.e. N saturation) that is  
3 derived from the FAB model. This approach requires the input of multiple ecosystem  
4 parameters. Its components are expressed by eq 1.

$$N_{eco} = fN_{upt} + N_{ret} + (1 - r)(N_{imm} + N_{den}) \quad (1)$$

7  
8 Where,

9  $N_{upt}$  = nitroge n uptake by the catchment

10  $N_{imm}$  = nitroge n immobilization by the catchment soil

11  $N_{den}$  = denitrification of nitroge n in the catchment,

12  $N_{ret}$  = in-lake retention of nitroge n

13  $f$  = forest cover in the catchment (dimensionless parameter)

14  $r$  = fraction lake/catchment ratio (dimensionless parameter)

15  
16 The second approach for estimating  $N_{eco}$  is to take the difference between N  
17 deposition and measured N leaching in a catchment as expressed by eq 2.

$$N_{eco} = DL(N) - N_{leach} \quad (2)$$

18  
19  
20 The site specific values of critical loads can be used to derive such a deposition  
21 loading, here called the deposition metric, which represents a group or percentage of  
22 water bodies that reach a specified ANC (or higher) in a given spatial area. For example,  
23 if it is desired that all water bodies reach a specified ANC, the allowable amount of  
24 deposition for all water bodies is equal to the lowest critical load the population of water  
25 bodies. Because the deposition metric represents a percentage of individual catchments  
26 from a population of water bodies, and not an individual catchment, the deposition metric  
27 is noted by the follow abbreviation  $DL_{\%ECO}$ .

28 As an example of the above approach, we evaluated the population of 169  
29 waterbodies in the Adirondacks used in the REA analysis. For each individual waterbody  
30 in the population the critical load at  $ANC_{lim} = 50 \mu eq/L$  was calculated using the two  
31 equations for deriving the  $N_{ECO}$  term (eq 2 and 3). The distribution of deposition loads

1 for the population was assessed and Table A.1.3-1 shows example for the percentage of  
 2 protection. The mean value for DL<sub>%ECO</sub> for the 169 water bodies is presented, as well as  
 3 the values for which 50, 75, 85, 95 and 100% of the water bodies in the population will  
 4 not exceed their critical load at ANC = 50 µeq/L. Note, only 32% of water bodies would  
 5 not exceed their critical load at ANC = 50 µeq/L for the mean value DL<sub>%ECO</sub> because  
 6 variability is high in the data set, therefore the mean can be problematic for areas with  
 7 high variability.

8

Table A.1.3-1. Example calculations for determining the percent of water bodies achieving target ANC levels. This example is based the population of DL <sub>ANClim</sub> for and ANC=50 for 169 catchments in the Adirondacks. These catchments occur across three categories of geologic sensitivity. We could separate the DL <sub>ANClim</sub> values into sensitivity categories (if info is available) and do the analysis for each category or calculate one DL <sub>ANClim</sub> for combined geologic categories. Units are in meq/m <sup>2</sup> /yr.						
	NHx dep	Neco (eq2)	DL <sub>%ECO</sub> (S+N) +Neco (eq 2)	Neco (eq3)	DL <sub>%ECO</sub> (S+N) +Neco (eq 3)	% of lakes within the population that have ANC ≥ 50µeq/L
Mean	20.40	19.19	162.36	63.95	207.55	31.7%
St dev	3.22	3.03	162.92	11.15	165.42	
St er	0.25	0.23	13.04	0.86	13.24	
Rank						
%tile						
50%			99.33		139.22	50%
75%			65.62		110.37	75%
85%			54.89		95.53	85%
95%			45.12		83.99	95%
100%			30.22		59.07	100%

9

10 The values for the maximum deposition N and S based on DL<sub>%ECO</sub> at ANC=50  
 11 using the two approaches for N<sub>ECO</sub> and protective of 95 and 50% of the population of  
 12 water bodies, are given in Table A.1.3-2.

13

14

1

**Table A-3 Values for N and S deposition tradeoff curves for ANC = 50, protecting 95 and 50% of the population, in Adirondacks case study area as illustrated on Fig 5.1 and Fig 5.2. Units are in meq/m<sup>2</sup>/yr unless noted otherwise.**

% protection		NHxdep	Neco	DL <sub>%ECO</sub> (max N)	DL <sub>%ECO</sub> (max S)	DL <sub>%ECO</sub> (max NOY)
95	Eq 2	20.4	19.19	45.12	25.93	141.96
50	Eq 2	20.4	19.19	99.33	80.14	78.9.3
95	Eq 3	20.4	63.75	83.99	20.04	63.59
50	Eq 3	20.4	63.75	139.22	75.27	118.82

2  
3  
4  
5

6 **A.1.4 Additional Analysis of Bedrock Geology as a basis for defining acid-sensitivity**  
7 **classes across the landscape**

8  
9

**APPROACH**

10 This analysis applied a methodology developed by Sullivan et al. (2007) for their  
11 exercise to test the hypotheses on watershed sensitivity to acidic deposition. Sullivan et  
12 al.’s exercise focused on streams in the Southern Appalachian Mountains region. The  
13 classification scheme was based on lithologic maps and the stream chemistry for more  
14 than 900 sites. Using logistic regression to model the presence of acid-sensitive  
15 waterbodies (expressed as ANC) with respect to lithology class, the authors found “four  
16 variables were highly significant in predicting the probability of occurrence of acid-  
17 sensitive sites (defined for this analysis as having ANC ≤ 20 µeq/L) in the Southern  
18 Appalachians using logistic regressions: % siliceous bedrock in watershed, % forested  
19 watershed, elevation, and watershed area”. (Sullivan et al., 2007)

20

21 Using stream and lake ANC data from the Shenandoah Mountains and the  
22 Adirondack Mountains, along with lithologic classifications, linked forested area,  
23 elevation and watershed area, this NOx SOx exercise used logistic regression to  
24 determine if a correlation exists between lithology and ANC that is similar to the  
25 correlation found by Sullivan et al. in the Southern Appalachians. If similar to Sullivan  
26 et al.’s findings, then the findings of this NOx SOx exercise will be extrapolated spatially  
27 to identify areas of the U.S. potentially sensitive to aquatic acidification.

1  
2 Phase I of this analysis applied the Sullivan et al. methodology separately to the two 2009 NO<sub>x</sub>-  
3 SO<sub>x</sub> Risk and Exposure Assessment (Shenandoah Mountains and the Adirondack Mountains  
4 Case Study Areas) and consists of 4 steps:

- 5
- 6 1. Site selection (i.e., lakes and streams)
- 7 2. Stream chemistry data acquisition and conversion to a spatial data layer
- 8 3. Data acquisition for % lithology classification of watershed (e.g., % siliceous bedrock),  
9 elevation, % forested area of watershed, and watershed area
- 10 4. Logistic regression modeling.

11  
12 The modeling results are compared to Sullivan's SAMI analysis and, if consistent,  
13 will be extrapolated to the region surrounding the case study area (Phase II) and to other  
14 regions of the U.S. (Phase III) upon EPA directive.

15  
16 The output desired for this Phase I exercise was a set of constants and coefficients  
17 that predict the probability that the acid-sensitive watersheds (having stream ANC either  
18  $< 0 \mu\text{eq/L}$  or between 0 and  $20 \mu\text{eq/L}$ ) in the selected study areas based upon the input  
19 variables (percent lithology classification in watershed, elevation, watershed size, and  
20 percent forest cover in watershed).

## 21 22 **PHASE I**

### 23 24 **Step 1 – Site Selection: Streams and lakes to be assessed**

25  
26 The Adirondack Case Study Area and the Shenandoah Case Study Area provide ideal  
27 areas to assess the risk to aquatic ecosystems from NO<sub>x</sub> and SO<sub>x</sub> acidifying deposition. Four  
28 main reasons support the selection of these two areas. First, both regions fall within the areas of  
29 the United States known to be sensitive to acidifying deposition because of a host of  
30 environmental factors that make these regions predisposed to acidification. Second, these areas  
31 are representative of other areas sensitive to acidification. Third, these regions have in the past  
32 and continue to experience substantial exposure to NO<sub>x</sub> and SO<sub>x</sub> air pollution. Fourth, these

1 areas have been extensively studied ... over the last 3 decades (see Section 4 of the ISA Report  
2 (US EPA 2008). (US EPA, 2009, REA, Appendix 4, Section 3.1)

3  
4 Freshwater surveys and monitoring in the eastern United States have been  
5 conducted by many programs since the mid-1980s, including the National Lake/Stream  
6 Surveys (NSWS), EPA's Environmental Monitoring and Assessment Program (EMAP),  
7 the Temporally Integrated Monitoring of Ecosystems (TIME) monitoring program  
8 (Stoddard, 1990), and Long-Term Monitoring (LTM) project (Ford et al., 1993; Stoddard  
9 et al, 1998). The purpose of these programs is to determine the current state and  
10 document the trends over time in surface water chemistry for regional populations of  
11 lakes or streams impacted by acidifying deposition. Based on extensive surveys and  
12 surface water data from these programs, it was determined that the most sensitive lakes  
13 and streams (i.e., ANC less than about 50 µeq/l) in the eastern US are found in New  
14 England, the Adirondack Mountains, the Appalachian Mountains (northern Appalachian  
15 Plateau and Ridge/Blue Ridge region), northern Florida, and the Upper Midwest. These  
16 areas are estimated to contain 95% of the lakes and 84% of the streams in the United  
17 States that have been anthropogenically acidified through deposition (see Annex 4.3.3.2  
18 of the ISA, US EPA 2008). (US EPA, 2009, REA, Appendix 4, Section 3.1)

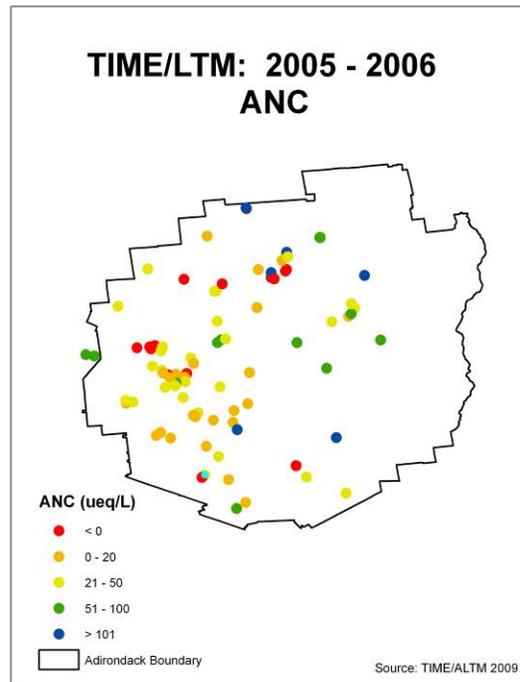
19  
20 ANC in surface water from 50 lakes in the Adirondack Case Study Area were monitored  
21 through the Adirondack LTM program and 38 lakes from the TIME program (**Figure A.1.4-3**).

22  
23 For the Adirondack Case Study Area, the regional EMAP probability survey of 117 lakes  
24 (i.e., weighting factors) were used to infer the number of lakes and percentage of lakes that  
25 receive acidifying deposition above their critical load of a target population of 1,842 lakes. The  
26 117 lakes...represent a subset of 344 sampled lakes throughout New England from 1991 through  
27 1994. (ME, NH, VT, RI, MA, CT, NY, NJ). (U.S. EPA, 2009, Appendix 4, Section 4.3.1.1)

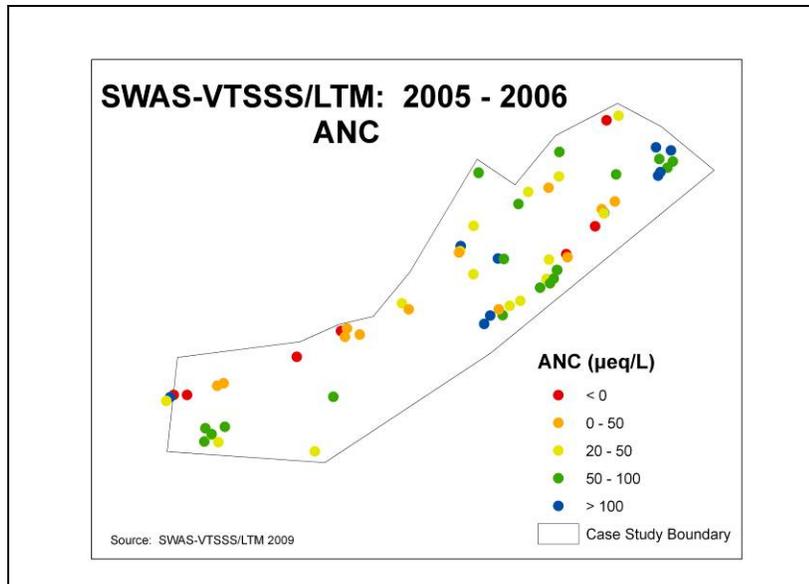
28  
29 Shenandoah Case Study Area ANC monitoring occurred as part of the  
30 Shenandoah National Park Surface Water Acidification Study (SWAS), Virginia Trout  
31 Stream Sensitivity Survey (VTSSS), and LTM programs. There are a significant number  
32 of the 67 streams in SWAS-VTSSS and LTM programs that currently have ANC of

1 about 50  $\mu\text{eq/L}$  based on the observed annual average ANC concentrations (**Figure**  
2 **A.1.4-4**).

3  
4 The total number of brook trout streams represented by the SWAS-VTSSS LTM  
5 quarterly monitored sites is approximately 310 out of 440 mountain headwater streams known to  
6 support reproducing brook trout in the Shenandoah Case study Area. ... The SWAS-VTSSS LTM  
7 programs began in Spring 1987, when water samples were collected from 440 streams known to  
8 have brook trout. Following the 1987 survey, a representative subset of 69 streams was selected  
9 for long-term quarterly monitoring of water quality, mostly located on National Forest lands or  
10 within the Shenandoah National Park Case Study Areas (14 SWAS and 55 VTSSS streams).  
11 (U.S. EPA, Appendix 4, Section 4.3.1.2).



12 **Figure A.1.4-3.** Current yearly average for 2005 to 2006 ANC ( $\mu\text{eq/L}$ ) in  
13 surface waters from 88 monitored lakes in the Temporally Integrated  
14 Monitoring of Ecosystems (38 Lakes) and Adirondacks Long-Term  
15 Monitoring (50 Lakes) networks in the Adirondack Case Study Area.  
16 (U.S. EPA, 2009)



1 **Figure A.1.4-4.** Current yearly average for 2005 to 2006 ANC ( $\mu\text{eq/L}$ ) in surface  
 2 waters from 67 monitored streams in the Surface Water Acidification Study,  
 3 Virginia Trout Stream Sensitivity, and Long-Term Monitoring network in the  
 4 Shenandoah Case Study Area.

5  
 6 [In Phase I, efforts focused on the Shenandoah and Adirondack Mountains case study  
 7 areas.]  
 8

9 **Step 2: Stream Chemistry Data Acquisition - Water chemistry for those streams and lakes**

10  
 11 There are numerous chemical constituents in surface water that can be use to indicate the  
 12 acidification condition of lakes and streams and to assess the effects of acidifying deposition on  
 13 ecosystem components. These include surface water pH and concentrations of  $\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$ ,  $\text{Al}_3^+$ ,  
 14 and  $\text{Ca}_2^+$ ; the sum of base cations; the recently developed base cation surplus; and the acid  
 15 neutralizing capacity (ANC). Each of these chemical indicators provides direct links to the health  
 16 of individual biota and the overall health and integrity of aquatic ecosystems as a result of surface  
 17 water acidification.

18  
 19 Although ANC does not directly affect the health of biotic communities, it is calculated  
 20 (or measured) based on the concentrations of chemical constituents that directly contribute to or  
 21 ameliorate acidity-related stress, in particular, pH,  $\text{Ca}_2^+$ , and dissolved inorganic aluminum.  
 22 Furthermore, numerical models of surface water acidification can more accurately estimate ANC  
 23 than all of the individual constituents that comprise it. Consequently, for the purpose of the  
 24 performing the case studies reported in the Risk and Exposure Assessment (US EPA, 2009),

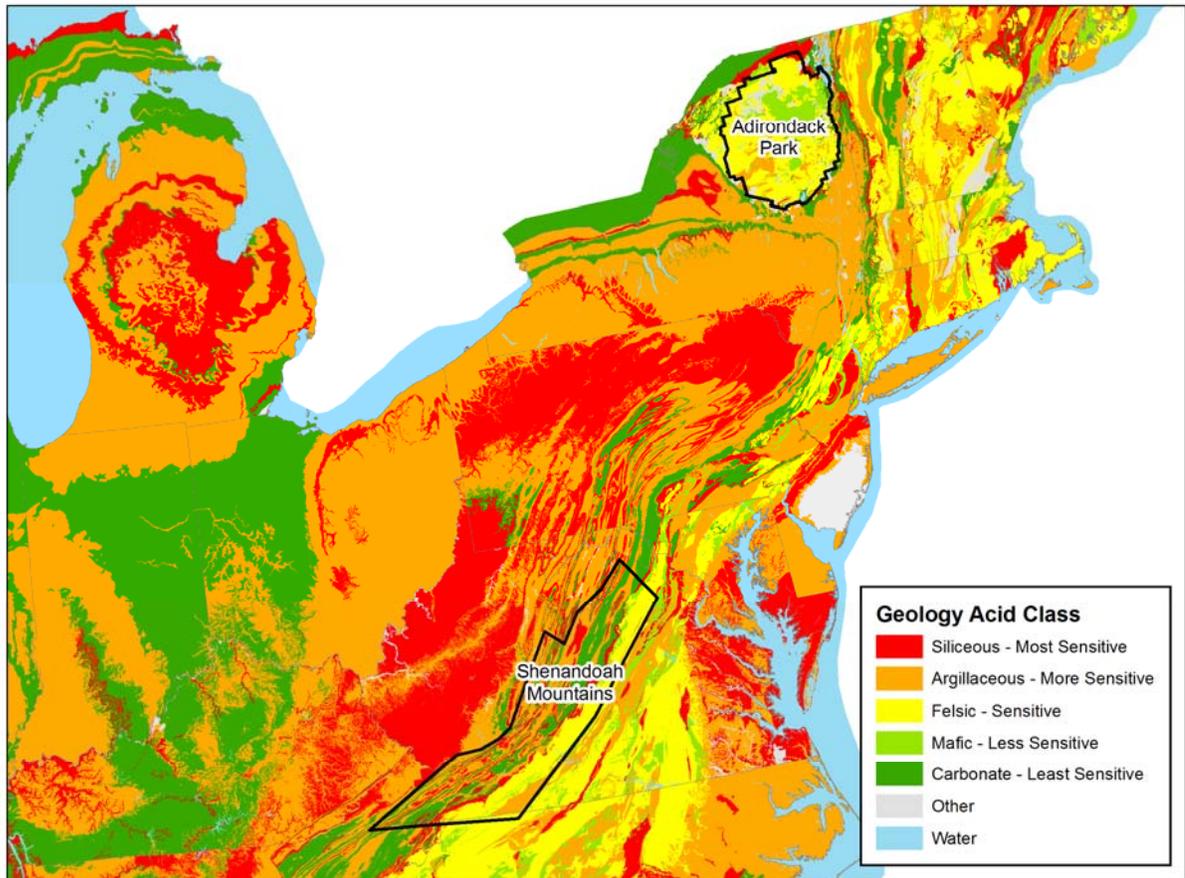
1 annual average ANC of surface waters was used as the primary metric to quantify the current  
2 acidic conditions and biological impacts for a subset of waterbodies in the study areas. (U.S.  
3 EPA, 2009 (REA, Appendix 4, Section 2.2)).  
4

5 All lake and stream monitoring data within the Adirondack and Shenandoah study  
6 areas were used.. Although Sullivan et al. used only Spring data because those values  
7 represented the time of year when ANC is at its lowest, and is also a time when sensitive  
8 life stages (eggs and young) are present for many fish species (Kaufman, Herlihy, Mitch,  
9 Messer,& Overton, 1991) this study used data throughout the calendar year. This was  
10 done because of the considerable variation within and between the study areas in terms of  
11 the time of lowest seasonal ANC, and the life cycles of the dependent biota. ANC  
12 values were then averaged by sampling station. Those stations in the Adirondacks and  
13 Shenandoah regions that fell within a 12 digit HUC that crossed into either of the study  
14 area boundaries were extracted to separate GIS data layers. These two collections of 12  
15 digit HUCs were then used to extract and average the other data parameters.

16  
17 **Step 3: Data acquisition for percent lithology classification, elevation, percent forested area**  
18 **of watershed, and watershed size**

19  
20 *Bedrock and surficial lithology* – Information on lithology for the eastern United States  
21 was been acquired from the states’ Geologic Surveys (USGS) and processed by EPA. This  
22 information was needed to determine if Sullivan et al.’s geologic classification system for the  
23 Southern Appalachian Mountains as applied to the eastern United States is a significant predictor  
24 of acid sensitivity in the Shenandoah Mountains and the Adirondack Mountains. Determining  
25 which acid class a given rock lithology falls into can be a somewhat subjective process. Upon  
26 initial inspection of the data, it appeared that there were some differences found between adjacent  
27 states (**Figure A.-1**). Following discussion with EPA, it was determined that the Adirondack  
28 data would benefit from re-evaluation of the classifications assigned. The acid sensitivity classes  
29 were reviewed for consistency and correctness and found to be accurate in all but a handful of  
30 cases. These exceptions were modified to match their lithologic descriptions. The Shenandoah  
31 class assignments were deemed acceptable. The percent of each of the five classes of acid  
32 sensitivity (siliceous, argillaceous, felsic, mafic, and carbonate) were tabulated over each of the  
33 12 digit HUCs that fell within the study areas.

1 **Figure A.1 Distribution of geology acid classes in the eastern United States and the**  
2 **location of the ELSDS4 sample locations.**



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**Elevation**, feet (30m Digital Elevation Maps) The water quality sample locations for the Adirondack and Shenandoah Mountains were extracted to a single shapefile, which was then overlaid with a national 30 meter digital elevation model (DEM). The elevation was determined at each location and extracted as an attribute.

**Watershed area**, km<sup>2</sup> (HUC12) The HUC 12 identifier was extracted using GIS in a manner similar to the extraction of elevation information for each acid-sensitive sample location. The area (km<sup>2</sup>) of the HUC 12 watershed was extracted to the water sampling location shapefile as an attribute.

**Percent forested area by watershed** has been estimated using 1992 US Forest Service vegetative databases and the Advanced Very High Resolution Radiometer (AVHRR) satellite

1 data (1,000 m cell size) to produce a raster forest type image with a 250 m cell size. This forest  
2 type image was processed using ArcGIS's Spatial Analyst by HUC 12 to determine the number  
3 of forested versus non-forested cells in each watershed. The results of this analysis were then  
4 used to calculate the percent of forested area in each watershed.

#### 6 **Step 4: Logistic Regression Modeling**

7  
8 Following Sullivan et al. (2007) a logistic regression approach with stepwise  
9 variable selection procedure was used to model binary variables derived from ANC. A  
10 significance level of  $\alpha=0.20$  was used as the criterion for variable entry, and  $\alpha=0.25$   
11 was selected as the criterion for remaining in the model (Hosmer and Lemeshow (1989),  
12 p.108). The following continuous predictor variables were considered in this modeling  
13 exercise : Elevation (ft), Area (km<sup>2</sup>), % Felsic, %\_Carbonate, %Mafic, %\_Siliceous, %  
14 Argillaceous, and %\_FOREST. SAS PROC Logistic (SAS (version 9.2)) was used to  
15 determine the best model.

16  
17 Three binary responses were considered in the logistic modeling:

- 18 1- ANC<=20 then outcome=1; else outcome=0
- 19 2- ANC<=50 then outcome=1; else outcome=0
- 20 3- ANC<=100 then outcome=1; else outcome=0.

#### 21 22 23 **ADDITIONAL INFORMATION ABOUT THE METHODOLOGY**

24  
25 The model was run for all sites' elevations and then followed by subsequent  
26 model runs that eliminate sites below a selected elevation and above a selected elevation  
27 to discern any improvement in correlation sensitive to elevation. The lower and upper  
28 elevations were selected based on the distribution of locations provided in Step 1.

#### 29 30 **EXTRAPOLATION – PHASES II AND III**

31  
32 The overall goal of this exercise was to develop constants and coefficients that  
33 predict the acid-sensitivity of watersheds with a high degree of probability. A different  
34 set of coefficients were developed for each distinct geographic region for which there is

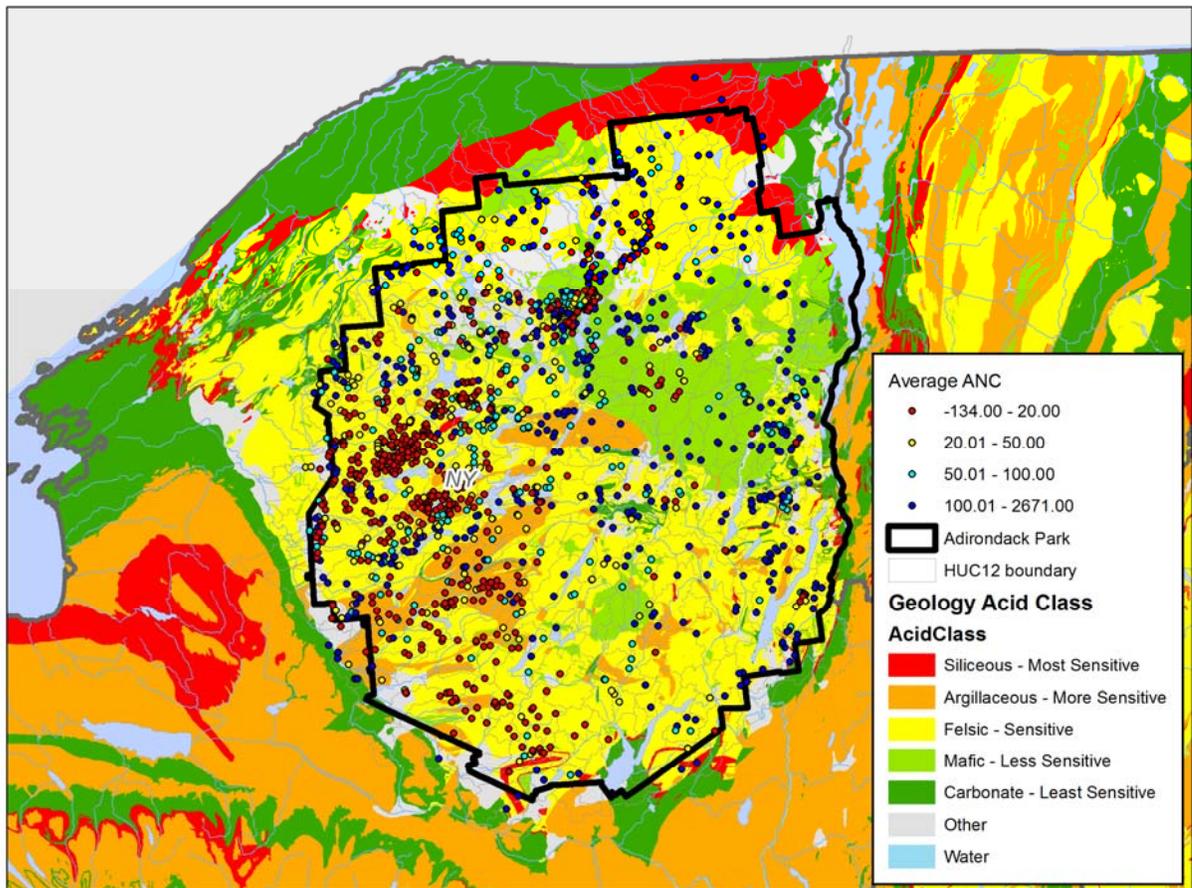
1 water quality monitoring data. In Phase II, the acid-sensitivity probability was calculated  
2 and mapped using GIS for all watersheds within each distinct geographic region of the  
3 case study areas, using the coefficients developed within the region.

4  
5 In Phase III, the acid-sensitive probability for watersheds in other regions outside  
6 the sampled regions would be calculated and mapped using GIS by applying the set of  
7 coefficients from the sampled region which is most similar, and for which all 4 types of  
8 input data are available (i.e., percent lithology classification of watershed, percent  
9 forested watershed, elevation, and watershed area). Classified lithology data are  
10 currently only compiled for the eastern half of the United States, so extrapolation will  
11 only be possible for this portion of the country at this time.

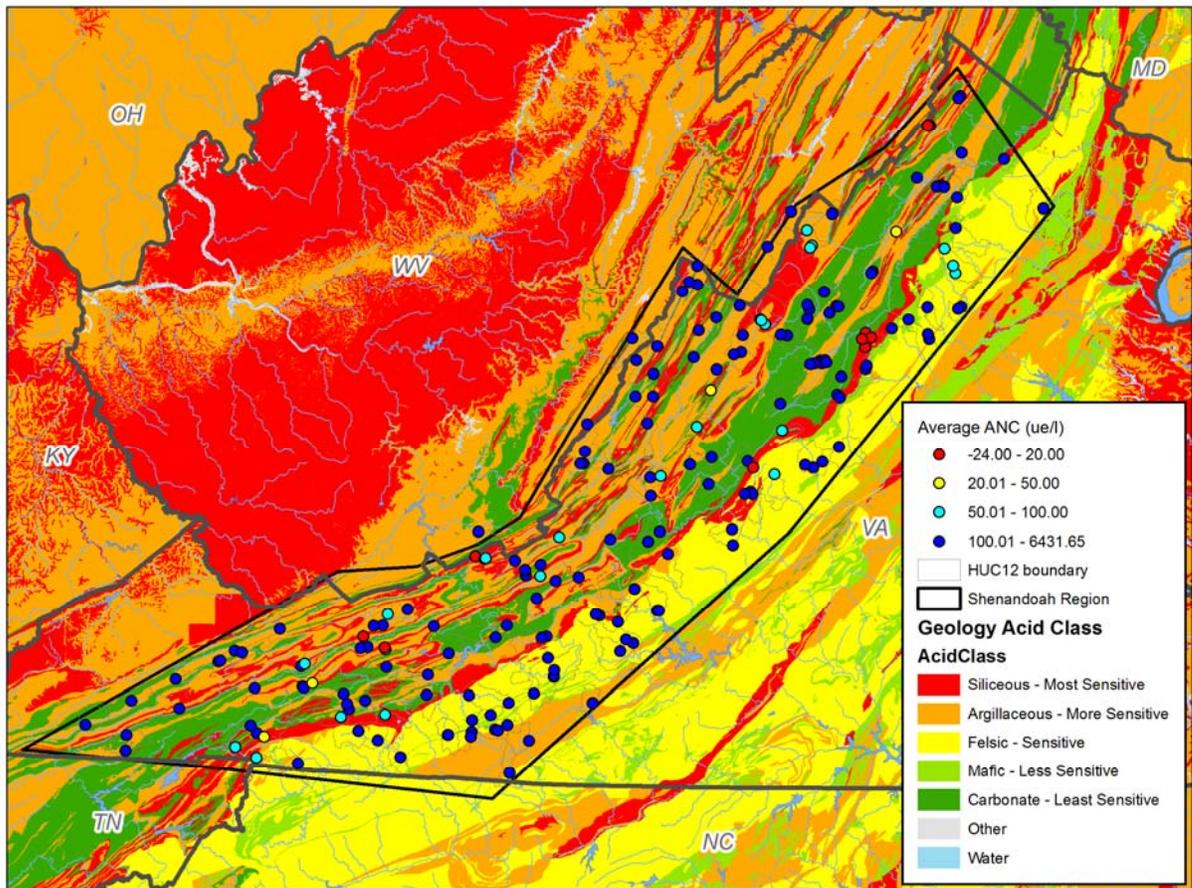
## 12 **RESULTS**

### 13 **GIS Mapping of Geologic Classifications and Measured ANC**

14  
15  
16  
17 Figures A-2 and A-3 show the distribution of water sample locations in the Adirondack  
18 and Shenandoah Mountains, respectively. The average ANC values are symbolized  
19 using 20, 50, and 100 microequivalents/litre as the break points. The five geology acid  
20 classes are also shown.



1  
2 **Figure A-2. Average ANC and Geology Classification: Adirondack Region**



1  
2 **Figure A-3. Average ANC and Geology Classification: Shenandoah Region**

3  
4 **Logistic Regression Analysis Results**

5  
6 Before running any modeling technique, the correlation between the predictors and ANC  
7 was explored (See Figure A.-4) A cell in the interception of a row and column represents  
8 the correlation between the variables at the end of the row and column. The correlation  
9 coefficient,  $R^2$ , ranges from -70 to 30%, suggesting a low to moderate correlation  
10 between predictors and outcome. Negative correlations indicate that as the predictor  
11 value increases, the value of ANC decreases. Positive correlations suggest that as the  
12 value of the predictor increases so does the value of ANC. The correlation matrix also  
13 serves to explore the multi-co-linearity issue (presence of highly correlated predictors).

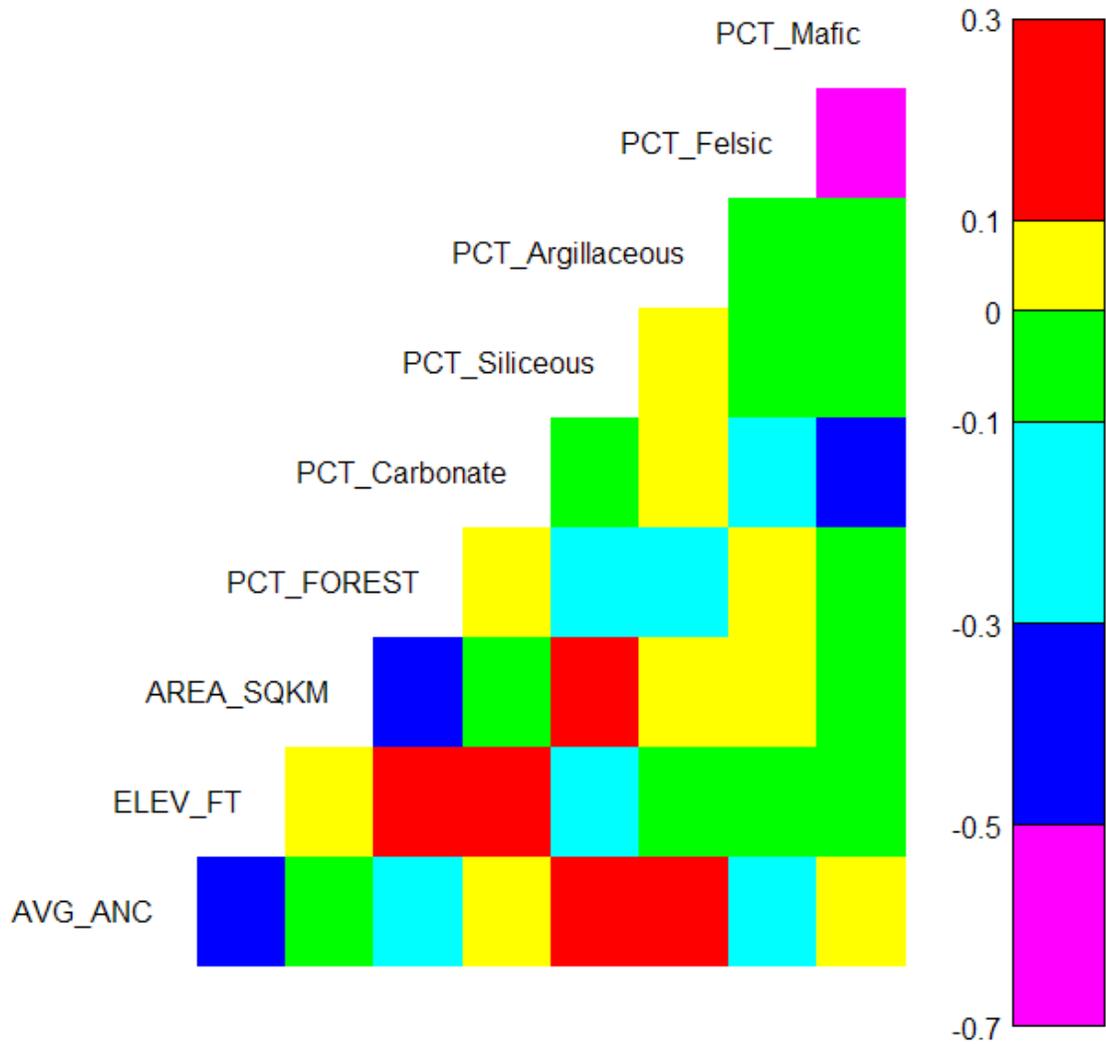
14  
15 In the Adirondacks region, Elevation is the variable with higher correlation (-30% to -  
16 50%), suggesting that this variable will likely be included as a predictor in the model.

17  
18 In the Shenandoah region, % Carbonate ( $R^2$  between 40% and 70%) and % Forest ( $R^2$   
19 between -40% to -70%) show a modest to high correlation with ANC. (See Figure A-5.)  
20 Also, % Forest is moderately correlated with % Siliceous ( $R^2$  between 40% to 70%) and

1 % Carbonate ( $R^2$  between -40% to -70%), suggesting that it is likely that one of these  
 2 three variables will be in the model. Similarly, % Felsic is moderately correlated with %  
 3 Carbonate and % Argillaceous ( $R^2$  between -40% to -70%) suggesting that if one of these  
 4 variables is in the model, then the other two won't be in the model.

5

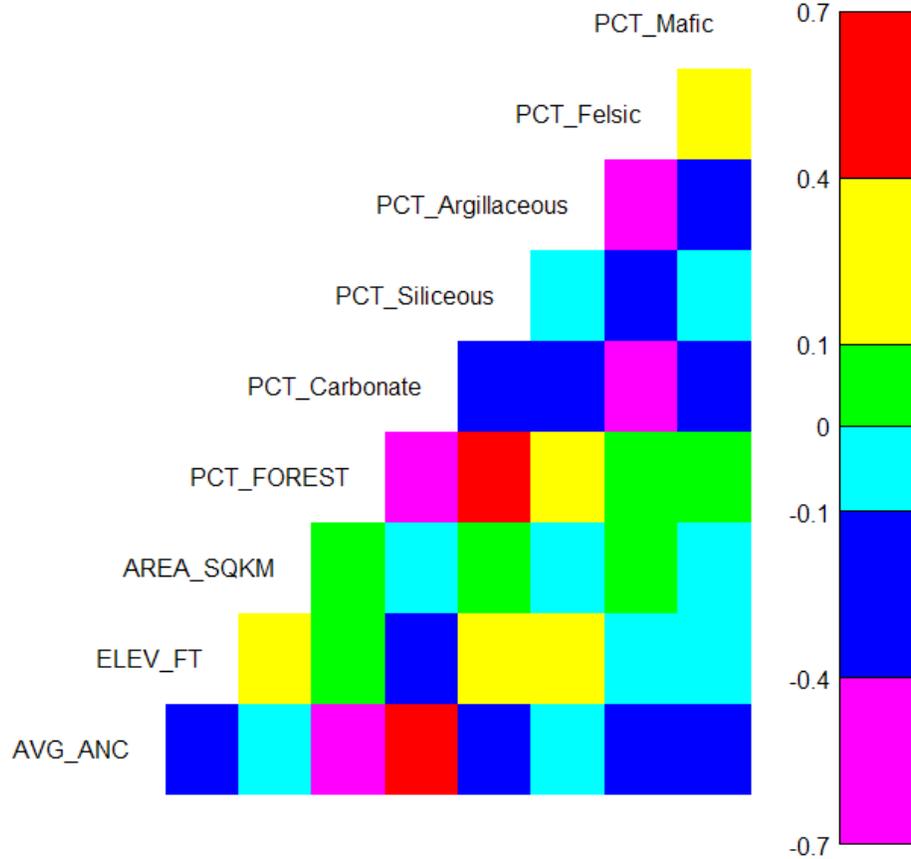
6 **Figure A-4. Correlation matrix for Adirondacks Region.**



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**Figure A-5. Correlation matrix for Shenandoah Region.**



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Among the measurements of model assessment considered were C statistic (a C value of 0.5 means the model predictions are equivalent to a random guess; >0.7 denotes worthwhile to the model); the Akaike information Criteria (AIC) (Hosmer and Lemeshow (1989), p.184); and the Schwarz information criterion (SC) (Kass, R. E. & Wasserman, L. (1995)); and the -2\*maximized log-likelihood (-2ML). Smaller values of all three statistics suggest a better model. To evaluate the goodness of fit of the model, the Hosmer and Lemeshow goodness of fit test (Hosmer and Lemeshow (1989)) was used (larger p-values suggest no difference between observed and model-predicted values, implying that the model's estimates fit the data at an acceptable level).

1 The final model for the Adirondacks Region binary variables is the following:

2

3 For the binary variable  $Y_{20} = \begin{cases} 1 & ANC \leq 20 \\ 0 & ANC > 20 \end{cases}$

4

5 the logistic model is the following:

6  $\text{logit}(p) = -3.3933 + 0.00305 \times \text{Elevation} - 0.0142 \times (\% \text{Forest}) - 0.0216 \times (\% \text{Carbonate})$   
 $- 0.0366 \times (\% \text{Siliceous}) - 0.0172 \times (\% \text{Mafic})$

7

8 For the binary variable  $Y_{50} = \begin{cases} 1 & ANC \leq 50 \\ 0 & ANC > 50 \end{cases}$

9

10 the logistic model is the following:

11  $\text{logit}(p) = -4.2271 + 0.00306 \text{Elevation} - 0.0197(\% \text{Carbonate}) - 0.0463(\% \text{Siliceous}) - 0.0155(\% \text{Mafic})$

12

13

14 For the binary variable  $Y_{100} = \begin{cases} 1 & ANC \leq 100 \\ 0 & ANC > 100 \end{cases}$

15

16 the logistic model is the following:

17

18

19 The final model for the Shenandoah region binary variables is the following:

20

21 For the binary variable  $Y_{20} = \begin{cases} 1 & ANC \leq 20 \\ 0 & ANC > 20 \end{cases}$

22

23 the logistic model is the following:

24  $\text{logit}(p) = -5.8725 + 0.0358(\% \text{Carbonate}) + 0.0632(\% \text{Siliceous})$

25 For the binary variable  $Y_{50} = \begin{cases} 1 & ANC \leq 50 \\ 0 & ANC > 50 \end{cases}$

26

27 the logistic model is the following:

28  $\text{logit}(p) = -6.0745 + 0.0233(\% \text{Carbonate}) + 0.0750(\% \text{Siliceous})$

29

30 For the binary variable  $Y_{100} = \begin{cases} 1 & ANC \leq 100 \\ 0 & ANC > 100 \end{cases}$

1  
 2 the logistic model is the following:  
 3  $\text{logit}(p) = -3.9068 + 0.000699(\% \text{Carbonate}) + 0.047(\% \text{Siliceous})$

4  
 5 Betas, standard error for betas, odds ratio and relevant statistics can be found in Appendix  
 6 2.

7  
 8 Weibull modeling using Proc Reliability (SAS) was also used to model ANC. LCL and  
 9 UCL denote the lower and upper confidence limits at 95% confidence level. If the value 0  
 10 is contained between the confidence limits then the parameter is not statistically  
 11 significant at 5% level.

12  
 13 **For the Adirondacks Region**, the final predictors and estimates are shown below:

14

15	Parameter	Estimate	SE	95% LCL
16	95%UCL			
17	Intercept	7.9493	0.3694	7.2254
18	ELEV_FT	-0.0015	0.0001	-0.0016
19	PCT_FOREST	-0.0085	0.0039	-0.0162
20	PCT_Carbonate	0.0021	0.0012	-0.0003
21	PCT_Argillaceous	0.0533	0.0208	0.0125

22  
 23 According to this model, ANC decreases with each unit increase in Elevation and Forest  
 24 (negative estimates) and increases with each unit increase in % Carbonate and %  
 25 Argillaceous (positive estimates).

26  
 27 Model assessment statistics (R2, AIC, etc.) were not found in the documentation. It is  
 28 possible to calculate them to evaluate the model fit to the data.

29  
 30 **For the Shenandoah Region**, the final predictors and estimates are shown below:

31

32	Intercept	7.3437	0.1585	7.0330	7.6543
33	ELEV_FT	-0.0015	0.0001	-0.0016	-0.0013
34	PCT_Siliceous	0.0167	0.0054	0.0061	0.0273
35	PCT_Argillaceous	0.0564	0.0209	0.0154	0.0973
36	PCT_Felsic	-0.0041	0.0011	-0.0062	-0.0020

37  
 38 According to this model, ANC decreases with each unit increase in Elevation and %  
 39 Felsic (negative estimates) and increases with each unit increase in % Siliceous and %  
 40 Argillaceous (positive estimates).

41  
 42 **Appendix 2 shows the odds ratio for each predictor.** The larger the Odds ratio, the  
 43 higher the chances that ANC <=20 for every change in unit of the predictor.

1 **For the Adirondacks region and ANC<=20**

2 When elevation increases one unit, the odds that ANC<=20 increase by a factor of 1.03,  
3 when other variables are controlled (meaning the same level of % Fforest, % Carbonate,  
4 % Siliceous and % Mafic). When % Forest increases one unit, then the odds that  
5 ANC<=20 decreases by a factor of 0.972 when other predictors are held constant. Similar  
6 interpretations exist for the rest of the models.

7  
8  
9 **For the Shenandoah region and ANC<=20:**

10  
11 When % Carbonate increases one unit, the odds that ANC<=20 increases by a  
12 factor of 1.036 when other predictors are held constant. Similarly, when % Siliceous  
13 increases one unit, the odds that ANC<=20 increases by a factor of 1.065 when other  
14 variables are held constant.

15  
16 **UNCERTAINTIES**

17  
18 *Lithology classification assignments.* RTI received a geodatabase of geology data  
19 from EPA to be used in the ANC GIS comparative analysis. The geodatabase includes a  
20 list of lithologies from the most recent USGS (Eastern Mineral Resources Team) national  
21 geologic map data for the continental United States. Each lithology polygon from the  
22 USGS map data set is defined by two lithology types labeled 'ROCKTYPE1' and  
23 'ROCKTYPE2'. According to the metadata for the geologic map cover, ROCKTYPE1  
24 characterizes more than 50% of the area in the associated polygon. EPA has categorized  
25 each lithology record into one of five geologic sensitivity classes (i.e., Siliceous,  
26 Argillaceous, Felsic, Mafic, and Carbonate), after Sullivan, et al (2006). The geologic  
27 sensitivity classification is included for each lithology record of the geodatabase in a field  
28 identified as 'AcidClassDesc'.

29 A cursory review was performed on EPA's assigned classifications suggesting--

- 30 • Not all rock types in the geodatabase are included in Sullivan (Table 1), but in  
31 most instances (except where noted) there was agreement between Sullivan's  
32 classification and the EPA assigned classes for rock types included in both the  
33 technical document and the geodatabase;

- 1       • Potential discrepancies in class assignments were identified such as those  
2       associated with non-descriptive lithologic description (e.g., sedimentary rock),  
3       others with typical rock chemistry and GSC (e.g., Carbonate GSC was assigned to  
4       beach sand), also classification assigned to sedimentary environments (e.g.,  
5       olistostrome), or terms describing non-mineralogy specific rock texture (e.g.,  
6       hornfel). The majority of these fell outside the Adirondack or Shenandoah study  
7       areas.

8

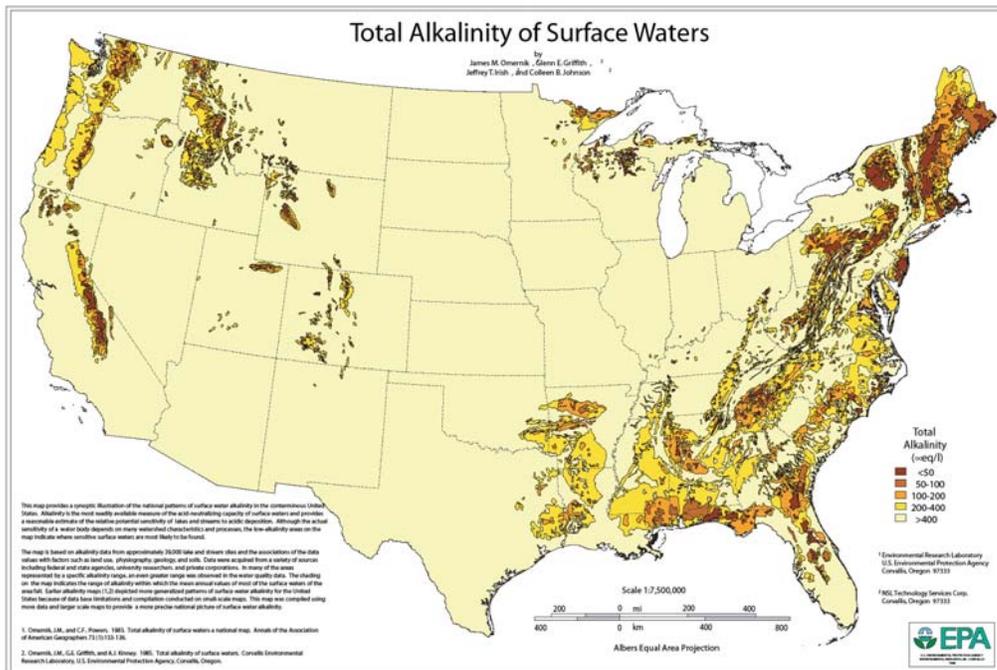
9       After discussion with EPA and a more detailed review, only a few revisions were  
10      required in the Adirondack datasets and those revisions were assigned less acid-sensitive  
11      classes. No revisions were required in the Shenandoah dataset.

12

1 **A.1.5 Additional techniques explored for which to base subdivision of the U.S. based based**  
2 **on acid sensitivity.**

3 ***Alkalinity***

4 We also considered dividing the U.S. based on the Alkalinity from the 1984  
5 Omernick Alkalinity map. We do not recommend using alkalinity because it includes  
6 more ecoregions as sensitive than the ANC approach, which causes the deposition loads  
7 for sensitive and less-sensitive areas to be more similar.



8 **Fig A.1.5-1. Omernick's alkalinity map (Omernick 1984)**

1

Table A.1.5-1. Descriptive statistics of the CL populations that result when the U.S. is divided into two categories, sensitive and non-sensitive based on Alk									
Omernick sensitivity classification									
CL for ANC =	Sensitivity class	CL in class n=	Mean Neco	Neco St er	CL mean (meq/m <sup>2</sup> /yr)	CL St er	Example deposition metrics		
							DL %90 (meq/m <sup>2</sup> /yr)	DL %75 (meq/m <sup>2</sup> /yr)	DL %50 (meq/m <sup>2</sup> /yr)
50	sensitive	5446	48	0.4	274	5	28	57	123
50	Non-sensitive	346	37	1	343	15	50	109	265

2

3 ***Cluster Analysis***

4 This approach uses ANC, ALK, DOC and soil BCw to categorize the acid-  
 5 sensitivity of ecoregions based on a quantitative cluster analysis. The ANC dataset is  
 6 described in Chapter 5, the ALK dataset is described in the previous section, and the  
 7 DOC and soil BC weathering datasets are described prior to the explanation of the cluster  
 8 analysis

9 ***DOC***

10 Water chemistry data on DOC was collected from several national monitoring  
 11 networks. The data sources are the same as those for ANC and summarized in Chapter  
 12 5 and include approximately 11,000 observations.

13

14 ***Bed rock geology***

15 Bedrock geology is the parent material to soils and can be related to acid-  
 16 sensitivity. In the first draft PA the EPA staff proposed using bedrock geology to inform  
 17 acid sensitivity categorization of the landscape. This approach entailed defining  
 18 categories of ecosystem sensitivity to acidification based on bedrock geology/ lithology.  
 19 The approach is supported by conclusions from the ISA in which geology is determined  
 20 to be the governing factor that drives ecosystem sensitivity to acidification (**ISA 3.2.4.1**).  
 21 A map was developed in this policy assessment to capture the heterogeneity of geologic  
 22 bedrock that occurs across the eastern U.S. and link it to ecosystem acid-sensitivity (Fig  
 23 A.1.5-2). The method is based on Sullivan et al. (2007) in which 70+ primary lithologies

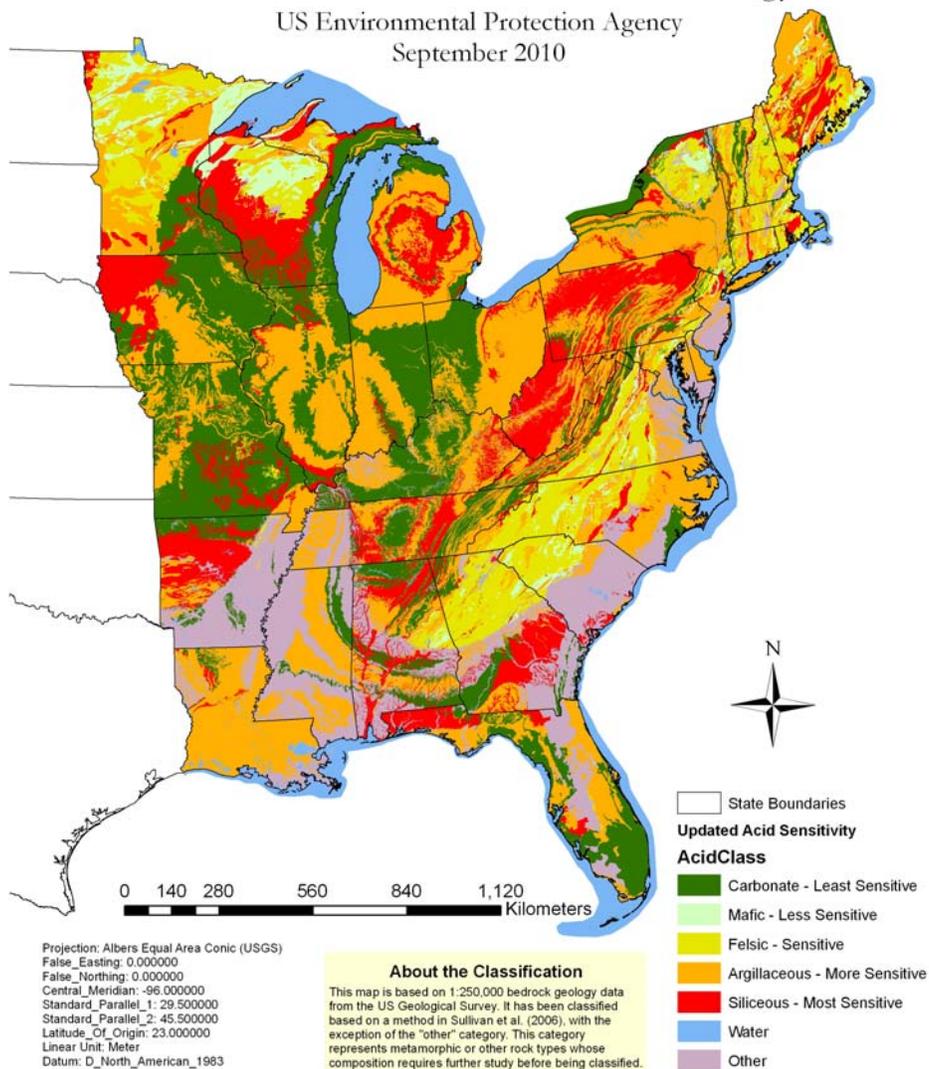
1 are grouped into 5 categories of acid-sensitivity, using ANC as the ecosystem indicator  
2 upon which acid-sensitivity is based. Sullivan et al. (2007) evaluated multiple features of  
3 the landscape and found that geology is the landscape parameter that governs ecosystem  
4 sensitivity to acidic deposition. The analysis in Sullivan et al. 2007 was conducted in the  
5 Southern Appalachian Mountains region, which included sites from the states of GA, TN,  
6 NC, KT, VA and WV. EPA conducted additional analyses to further test the concept that  
7 lithology correlates to acid sensitivity based on ANC, using data from the Adirondacks  
8 and Shenandoahs. Results from this analysis indicated that North of the glaciation line,  
9 bedrock geology was not a good predictor of acid sensitivity. For example, the  
10 Adirondacks which are known to be one of the most sensitive areas to acidification is  
11 dominated by types of bedrock geology that are considered moderately to less sensitive in  
12 areas South of the glaciation line.  
13

# Acid-Sensitive Areas of the Eastern United States

A Classification based on Bedrock Geology

US Environmental Protection Agency

September 2010



1

**Fig A.1.5-2. A map of acid sensitive areas of the Eastern U.S. developed from a lithology-based five-unit geologic classification system after methods in Sullivan et al. (2007).**

2

3

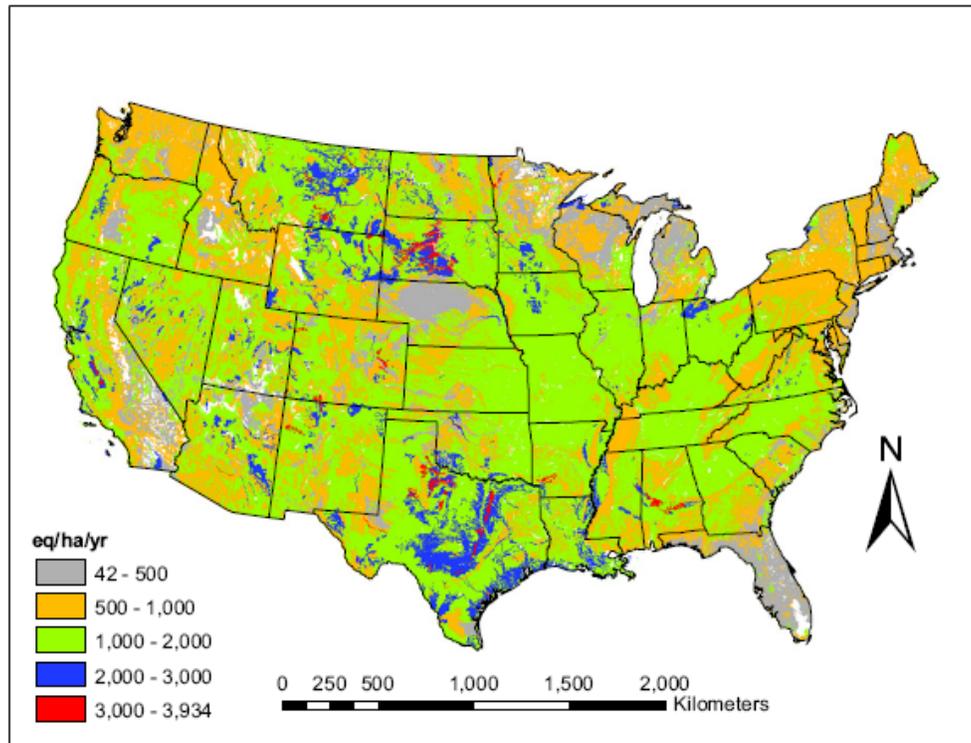
4

## 5 Soil base-cation weathering

6 As previously noted, the use of bedrock geology for the classification of acid sensitive  
7 catchments is problematic in areas north of the glaciation line because glacial activity has  
8 caused the surface till to become spatially disconnected from the parent geologic material.  
9 Surficial geology is therefore important to the evaluation of the spatial variability of acid-

1 sensitivity. For that reason a national map for soil base-cation weathering (McNulty et al.  
2 2007) was considered in the analysis of acid-sensitivity (Fig A.1.5-3).

3



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9

**Fig A.1.5-3.** A map of average annual forest soil base cation weathering expressed in  $\text{eq ha}^{-1} \text{yr}^{-1}$  for the conterminous US for the years 1994 to 2000 at a  $1\text{-km}^2$  spatial resolution from McNulty et al.

10

McNulty et al. 2007 estimated the base cation weathering rate using the clay correlation-substrate method (Sverdrup et al., 1990). This method used a combination of parent material and clay percent to determine the weathering rate. Base cation weathering rate ( $\text{eq ha}^{-1} \text{yr}^{-1}$ ). A temperature correction can be applied to this method, but this correction is more suitable for northern climates and was not used in the model. Clay fraction was derived from a weighted average of soil fraction per map unit.

15

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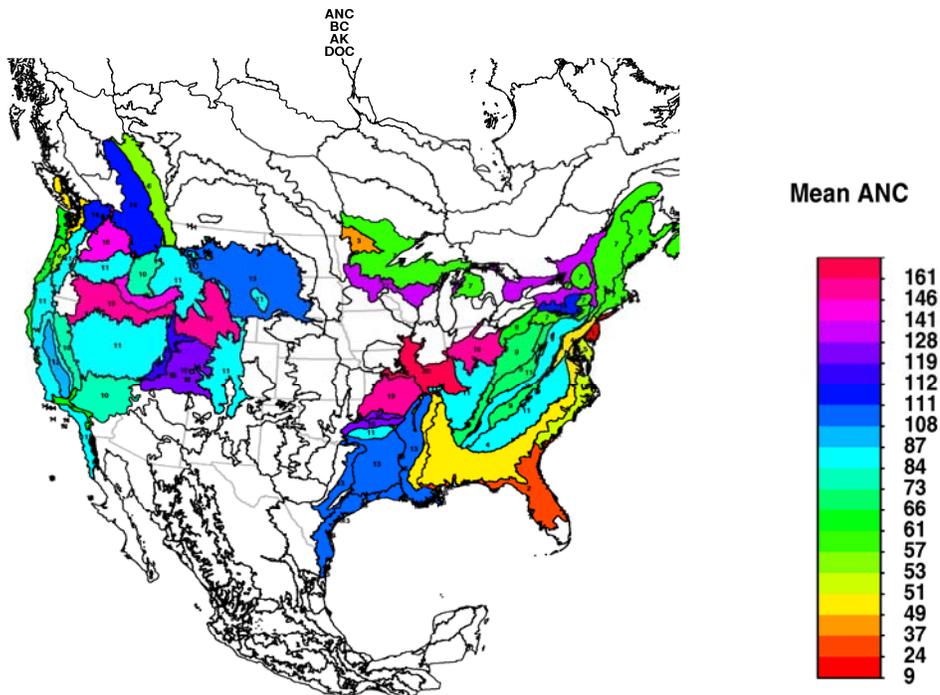
<b>Table A.1.5-3. Data input to the U.S. national soil base cation weathering map developed by McNulty et al. 2007</b>	
<i>Data source</i>	<i>Description of dataset</i>
State Soil Geographic Data Base (STATSGO)	Compilation by the Natural Resource Conservation Service (NRCS) of geology, topography, vegetation, climate, Landsat Thematic Mapper (TM) satellite imagery, and detailed, county level soil survey data. Each soil map unit in STATSGO includes multiple components and data layers (USDA NRCS, 1995). STATSGO is organized by state and consists of one geospatial vector representing the soil map units for that state and 15 tables describing characteristics of those map units. Multiple soil layers are associated with each map unit. Soil layer sampling depth is not consistent within a state or between states
CONUS-SOIL developed by the Earth System Science Center (ESSC) at Pennsylvania State University	This is a 1-km <sup>2</sup> multi-layer soil data set based on the STATSGO. ESSC converted the vector map unit layer in STATSGO to a 1-km <sup>2</sup> grid, remapped many of the original STATSGO attribute layers, and defined 11 standard soil layers (Miller and White, 1998). These data layers and tables linked the standardized data set to the original STATSGO data set distributed by ESSC as 1-km <sup>2</sup> soil map unit grids for the conterminous US. The CONUS-SOIL was much better suited for national-scale modeling than the original STATSGO attribute layers and was therefore used McNulty et al. 2007. Key soil data inputs included CONUS-SOIL map units and clay fraction (Miller and White, 1998).
forest soil percent organic matter (OM) layer	Was created by McNulty et al. 2007 by first averaging the maximum and minimum recorded values for OM were averaged for each layer in the STATSGO data set. Second, the average OM layers were remapped into the 11 standard CONUS-SOIL layers using a weighted average to redistribute the average OM STATSGO layers into the CONUS-SOIL layers. If a STATSGO layer was completely contained in a CONUS-SOIL layer, then the average OM was multiplied by the component percent to determine the average OM contribution to the standard layer. If the STATSGO layer overlapped more than one CONUS-SOIL layer, then the proportion of overlap was multiplied by the average OM and the component percent, where the component percent was the proportion of the soil map unit comprised of that soil component. Once the conversion from STATSGO to CONUS-SOIL layer was complete, the 11 standard layers were summed by layer and divided by the sum of component percent. Finally, the weighted average was calculated according to equation 8.
parent material class	was derived from the STATSGO map unit component (comp) and taxonomic (tax) classification tables (USDA NRCS, 1995). The dominant mineralogy for each soil map unit was determined from the comp and tax tables. Each unit was classified into parent material class based on the mineralogical description (USDA NRCS, 2006)
Soil depth	in meters was obtained from the CONUS-SOIL depth to bedrock layer. This layer identified map units with bedrock less than 1.52 m below the soil surface (i.e., map units coded with a depth of 1.52 m did not encounter bedrock) (Miller and White, 1998).

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**Results**

This approach uses ANC, ALK, DOC and soil BCw to categorize the acid-sensitivity of ecoregions based on a quantitative cluster analysis. Cluster analysis is a method to sort a set of observations (in this case CL from watersheds) into subgroups so that the degree of association between observations is maximal if they belong to the same group and

1 minimal otherwise. The similarity of each site was determined using ANC, ALK, DOC  
2 and BCw. In the cluster analysis, the *k*-means algorithm assigned each point to the cluster  
3 whose center (also called centroid) is nearest. The center is the average of all the points in  
4 the cluster — that is, its coordinates are the arithmetic mean for each dimension  
5 separately over all the points in the cluster. The ecoregions was then assigned  
6 membership to a cluster. The weakness of this approach is that each of the datesets for  
7 the criteria (ANC, soilBCw, Alk and DOC), have varying levels of national coverage. If  
8 not all four criteria are available for the analysis the ecoregions drops out and is not  
9 assigned a cluster identification. The majority of ecoregions in the U.S. were not  
10 represented in the datasets for all four criteria and were not assigned a cluster  
11 identification (Fig A.1.5-4). Therefore the ulitility of this approach is limited.  
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16

1 **APPENDIX B**

2 ***Analysis of Critical Loads, Comparing Aquatic and Terrestrial Acidification***

3  
4 *Background*

5 Critical load is defined as, “a quantitative estimate of ecosystem exposure to one or more  
6 pollutants below which significant harmful effects on specified sensitive elements of the  
7 environment do not occur, according to present knowledge” (McNulty et al., 2007), and critical  
8 loads can be estimated for aquatic and terrestrial ecosystems. Within the *Risk and Exposure*  
9 *Assessment for Review of the Secondary National Ambient Air Quality Standards for Oxides of*  
10 *Nitrogen and Sulfur* (hereafter referred to as REA Report) (US EPA, 2009), critical loads of  
11 acidification for aquatic systems were determined by relating specific amounts of acidifying  
12 nitrogen and sulfur deposition to selected Acid Neutralizing Capacities (ANC) within freshwater  
13 lakes or streams. The presence and abundance of fish species served as the biological indicator  
14 of the impacts of the exceedance of critical acid loads by nitrogen and sulfur deposition.  
15 Estimation of critical acid loads for terrestrial systems within the REA Report (US EPA, 2009)  
16 related acidifying nitrogen and sulfur deposition to the base cation to aluminum (Bc/Al) ratio in  
17 the soil solution, and the health of sugar maple and red spruce in forest ecosystems served as the  
18 biological indicator of the impacts of critical acid load exceedance. A main distinction between  
19 these two critical loads is that aquatic critical loads are largely an integrated function of the  
20 chemistry of run-off waters that feed the lake or stream within a watershed, while terrestrial  
21 critical acid loads are determined by the rooting zone section of the soil profile in a forest  
22 ecosystem. Therefore, it is possible to have different critical load values for aquatic and  
23 terrestrial ecosystems within the same watershed.

24  
25 The goal of this Task was to determine the relative degree of protection offered by  
26 aquatic versus terrestrial critical acid loads within a landscape. Critical acid loads for lakes and  
27 streams within watersheds of the Adirondacks and Shenandoah Valley were compared against  
28 terrestrial critical loads calculated for same watersheds to determine which estimate had the  
29 lowest, most protective critical load for acidifying nitrogen and sulfur deposition.

1 Methods

2 For the REA Report (US EPA, 2009), critical acid loads were determined for 169 lakes  
3 and 60 streams in the Adirondacks and Shenandoah Case Study Areas, respectively. These  
4 critical loads were calculated using four different ANCs, 0, 20, 50 and 100  $\mu\text{eq/L}$ , that ranged in  
5 the level of protection offered to fish species abundance and diversity, and the resulting critical  
6 acid loads were classified into four “current condition of acidity and sensitivity to acidification”  
7 categories. “Highly Sensitive” water bodies had critical loads less than or equal to 50  
8  $\text{meq/m}^2/\text{yr}$ , “Moderately Sensitive” systems had critical loads ranging from 51 to 100  $\text{meq/m}^2/\text{yr}$ ,  
9 “Low Sensitivity” lakes and streams had critical loads that ranged from 101 to 200  $\text{meq/m}^2/\text{yr}$ ,  
10 and “Not Sensitive” systems had critical acid loads greater than 201  $\text{meq/m}^2/\text{yr}$ .

11  
12 For the purposes of this Task, aquatic critical acid loads corresponding to an ANC of 50  
13  $\text{meq/m}^2/\text{yr}$  were selected, and the locations of the lakes and streams in the Adirondacks and  
14 Shenandoah Case Study Areas were mapped by HUC12 watersheds. Availability of data for  
15 terrestrial acidification estimates was determined for each HUC, and only HUCs that had  
16 sufficient data were mapped. Data from the U.S. Department of Agriculture- Natural Resources  
17 Conservation Service (USDA-NRCS) SSURGO soils database (USDA-NRCS, 2008) had the  
18 poorest coverage. This data restriction limited the number of water bodies that could be included  
19 in the analysis to 62 and 35 for the Adirondacks and Shenandoah Case Study Areas, respectively.

20  
21 To examine a representative selection of water bodies in each Case Study Area, four  
22 watersheds containing lakes or streams from each of the four “current condition of acidity and  
23 sensitivity to acidification” categories were randomly selected. Therefore, a total of 16  
24 watersheds were chosen for each Case Study Area. All four “current condition of acidity and  
25 sensitivity to acidification” categories were evenly represented for the Adirondacks Case Study  
26 Area (four watersheds for each of the four categories). However, due to the limited number of  
27 watersheds in the Shenandoah Area and a lower proportion of lakes with low sensitivities to  
28 acidifying nitrogen and sulfur deposition (“Low Sensitivity” and “Not Sensitive”), it was not  
29 possible to have equal representation of all “current condition of acidity and sensitivity to  
30 acidification” categories. Therefore, there was a larger representation of streams that were more  
31 sensitive to acidification (“Highly Sensitive” and “Moderately Sensitive”). All water bodies that

1 were located in each of the selected HUCs were included in the analyses. In many cases, these  
2 water bodies ranged in sensitivity to acidification. In total, 29 lakes and 20 streams were  
3 analyzed in the Adirondacks and Shenandoah Case Study Areas, respectively (**Table 1** and **2**).

- 1 **Table 1.** Watersheds (HUC 12) and fresh water lakes in the Adirondacks Case Study Area that were used in the comparison of
- 2 aquatic and terrestrial critical acid loads. Lake IDs and associated aquatic critical acid loads (CL) in meq/m<sup>2</sup>/yr, based on an ANC of
- 3 50 µeq/L, are indicated in each cell and are from the REA REPORT (US EPA, 2009).

HUC	CURRENT CONDITION OF ACIDITY AND SENSITIVITY TO ACIDIFICATION CATEGORY			
	Highly Sensitive (CL ≤ 50 meq/m <sup>2</sup> /yr)	Moderately Sensitive (CL = 51-100 meq/m <sup>2</sup> /yr)	Low Sensitivity (CL = 101-200 meq/m <sup>2</sup> /yr)	Not Sensitive (CL > 201 meq/m <sup>2</sup> /yr)
<b>020100010103</b>				NY534L (CL = 1043)
<b>020100040203</b>			1A2-028O (CL = 106) NY310L (CL = 147)	NY308L (CL = 485)
<b>020100080304</b>				NY312L (CL = 588) NY313L (CL = 598)
<b>020100081602</b>				NY500L (CL = 610)
<b>020200020101</b>		NY013L (CL = 64)		
<b>020200020704</b>		NY536L (CL = 69)		
<b>020200040805</b>	1A2-078O (CL = 33)		NY292L (CL = 117)	
<b>041501011001</b>	NY029L (CL = 39)			
<b>041503020801</b>				NY783L (CL = 455)
<b>041503040102</b>	NY284L (CL = 23) NY285L (CL = 42)			
<b>041503040204</b>		NY278L (CL = 57)		
<b>041503050103</b>	1A1-089O (CL = 43)	050215AO (CL = 74) NY793L (CL = 97)		
<b>041503050104</b>	NY290L (CL = 30) NY289L (CL = 50)			

HUC	CURRENT CONDITION OF ACIDITY AND SENSITIVITY TO ACIDIFICATION CATEGORY			
	Highly Sensitive (CL $\leq$ 50 meq/m <sup>2</sup> /yr)	Moderately Sensitive (CL = 51-100 meq/m <sup>2</sup> /yr)	Low Sensitivity (CL = 101-200 meq/m <sup>2</sup> /yr)	Not Sensitive (CL > 201 meq/m <sup>2</sup> /yr)
041503050302			NY008L (CL = 146) NY007L (CL = 165)	
041503050407		NY767L (CL = 51) NY529L (CL = 73) NY528L (CL = 82) NY769L (CL = 99)	NY768L (CL = 114)	
041503050601			NY004L (CL = 168)	

1

1 **Table 2.** Watersheds (HUC 12) and streams in the Shenandoah Case Study Area that were used in the comparison of aquatic and  
 2 terrestrial critical acid loads. Stream IDs and associated aquatic critical acid loads (CL) in meq/m<sup>2</sup>/yr, based on an ANC of 50 µeq/L,  
 3 are indicated in each cell and are from the REA REPORT (US EPA, 2009).  
 4

HUC	CURRENT CONDITION OF ACIDITY AND SENSITIVITY TO ACIDIFICATION CATEGORY			
	Highly Sensitive (CL ≤ 50 meq/m <sup>2</sup> /yr)	Moderately Sensitive (CL = 51-100 meq/m <sup>2</sup> /yr)	Low Sensitivity (CL = 101-200 meq/m <sup>2</sup> /yr)	Not Sensitive (CL > 201 meq/m <sup>2</sup> /yr)
020700050401	VT37 (CL = 26)			
020700050502	VT57 (CL = 39)			
020700050703	VT40 (CL = 13)			
020700050705	VT35 (CL = 37) VT36 (CL = 24)			
020700050801	DR01 (CL = 33) WOR1 (CL = 43)			
020700050803	VT53 (CL = 40)			
020700060101		VT54 (CL = 69)		
020801030301			VT60 (CL = 198)	VT61 (CL = 231)
020801030402		VT62 (CL = 68)		
020802010702	VT10 (CL = 15)			
020802010703	VT11 (CL = 14)	VT12 (C = 75)		
020802010801	VT14 (CL = 14) VT15 (CL = 13)			
020802010803	VT16 (CL = 20)			
020802020102		VT38 (CL = 66)		
020802020401	VT41 (CL = 15)			
020802030601		VT46 (CL = 52)		

1 Terrestrial critical acid loads were calculated for each of the 16 watersheds using the  
 2 simple mass balance method (UNECE, 2004) and data sources outlined in the REA Report (US  
 3 EPA, 2009), and Bc/Al soil solution indicator values of 1.2 and 10.0. Briefly, average values for  
 4 base cation deposition (calcium, potassium, magnesium and sodium), chloride deposition, and  
 5 annual runoff ( $\text{m}^3/\text{ha}/\text{yr}$ ) were determined for each watershed (**Table 3**). The Kgibb constant  
 6 ( $\text{m}^6\text{eq}^2$ ) was determined by the average percent organic matter in the soil (**Table 4**), and N  
 7 immobilization in the soil was set to the constant value of 42.86 eq/ha/yr (McNulty et al., 2007).  
 8 It was assumed that active harvesting did not occur in each of the watersheds. Therefore base  
 9 cation (calcium, magnesium and potassium) and nitrogen uptake were 0 eq/ha/yr (UNECE,  
 10 2004). Similarly, it was assumed that the majority of each watershed consisted of upland sites.  
 11 Therefore, denitrification losses were assumed to be 0 eq/ha/yr (McNulty et al., 2007). Base  
 12 cation weathering was estimated using the clay substrate model (equations 1-3) (McNulty et al.,  
 13 2007).

$$14 \quad \text{Acid Substrate: } BC_e = (56.7 \times \% \text{clay}) - (0.32 \times (\% \text{clay})^2) \quad (1)$$

$$15 \quad \text{Intermediate Substrate: } BC_e = 500 + (53.6 \times \% \text{clay}) - (0.18 \times (\% \text{clay})^2) \quad (2)$$

$$16 \quad \text{Basic Substrate: } BC_e = 500 + (59.2 \times \% \text{clay}) \quad (3)$$

17 where

18  $BC_e$  = empirical soil base cation ( $\text{Ca}^{2+} + \text{K}^+ + \text{Mg}^{2+} + \text{Na}^+$ ) weathering rate  
 19 (eq/ha/yr)

20 % clay = the percentage of clay within the top 50cm of the soil.

21

22 The U.S. Department of Agriculture- Natural Resources Conservation Service (USDA-NRCS)  
 23 SSURGO soils database (USDA-NRCS, 2008)) and state-level geology (U.S. Geological Survey  
 24 (USGS) state-level integrated map database for the United States (USGS, 2009)) were used to  
 25 determine parent material acidity classification. Parent material acidity was determined for each  
 26 SSURGO polygon within each watershed using the criteria outlined in the REA Report (US  
 27 EPA, 2009), and the contributions of base cations from the weathering of acid, intermediate and  
 28 basic substrates (eq/ha/yr) were determined by a weighted average based on the proportion of  
 29 area occupied by each parent material acidity class. Rooting depth was assumed to be 50 cm and  
 30 masses of calcium, magnesium, potassium, sodium and nitrogen were converted to eq/ha/yr units

1 based on molar charge equivalents. Unless indicated otherwise, the units used in the calculation  
 2 of critical acid loads were eq/ha/yr. The estimated terrestrial critical loads for the 16 watersheds  
 3 in the Adirondacks and Shenandoah Case Study Areas are presented in **Table 5**.

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**Table 3.** Name, type and source of data used in the simple mass balance estimates of terrestrial critical acid loads for the watersheds in the Adirondacks and Shenandoah Case Study Areas.

DATA	NAME	TYPE	SOURCE
Base cation (Ca <sup>2+</sup> , Mg <sup>2+</sup> , Na <sup>+</sup> , K <sup>+</sup> ) deposition— wet	CMAQ/ NADP	GIS datalayers	Provided by U.S. Environmental Protection Agency (EPA)/NADP, 2003a,c, d, e
Chloride (Cl <sup>-</sup> ) deposition— wet	NADP	GIS datalayer	NADP, 2003b
Runoff	Annual run-off (1: 7,500,000 scale)	GIS datalayer	Gebert et al., 1987
Soil horizon depth	SSURGO	GIS datalayer	USDA-NRCS, 2008
Percentage of clay by soil horizon	SSURGO	GIS datalayer	USDA-NRCS, 2008
Percentage of organic matter by soil horizon	SSURGO	GIS datalayer	USDA-NRCS, 2008
Soil parent material	SSURGO	GIS datalayer	USDA-NRCS, 2008
State-level bedrock geology	State Geological Map Compilation	GIS datalayer	USGS, 2009

**Note:** CMAQ = Community Multiscale Air Quality Model; NADP = National Atmospheric Deposition Program; GIS = Geographic Information System; SSURGO = Soil Survey Geographic Database

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- 1 **Table 4.** Gibbsite equilibrium ( $K_{gibb}$ ) constant determined by percentage of soil organic matter  
 2 (modified from McNulty et al. 2007).

Soil Type Layer	Organic Matter %	$K_{gibb}$ ( $m^6/eq^2$ )
Mineral soils: C layer	<5	950
Soils with low organic matter: B/C layers	5 to 15	300
Soils with some organic material: A/E layers	15 to 70	100
Peaty and organic soils: organic layers	>70	9.5

3

1 **Table 5.** Terrestrial critical acid loads (in eq/ha/yr) for the watersheds in the Adirondacks and  
 2 Shenandoah Case Study Areas.  
 3

Case Study Area	HUC12	Terrestrial Critical Acid Load (eq/ha/yr)	
		Bc/Al = 1.2	Bc/Al = 10.0
Adirondacks	020100010103	2045	1134
Adirondacks	020100040203	1316	712
Adirondacks	020100080304	1329	731
Adirondacks	020100081602	1670	922
Adirondacks	020200020101	1484	819
Adirondacks	020200020704	1707	935
Adirondacks	020200040805	1770	951
Adirondacks	041501011001	1770	955
Adirondacks	041503020801	1664	912
Adirondacks	041503040102	1627	880
Adirondacks	041503040204	1436	786
Adirondacks	041503050103	1774	957
Adirondacks	041503050104	1794	968
Adirondacks	041503050302	1754	947
Adirondacks	041503050407	1447	789
Adirondacks	041503050601	1203	656
Shenandoah	020700050401	1440	802
Shenandoah	020700050502	1560	871
Shenandoah	020700050703	1762	979
Shenandoah	020700050705	1852	1032
Shenandoah	020700050801	1799	1003
Shenandoah	020700050803	1975	1102
Shenandoah	020700060101	1638	914
Shenandoah	020801030301	1511	843
Shenandoah	020801030402	1393	776
Shenandoah	020802010702	1603	890
Shenandoah	020802010703	1642	912
Shenandoah	020802010801	1635	909
Shenandoah	020802010803	1573	876
Shenandoah	020802020102	1519	845
Shenandoah	020802020401	1264	703
Shenandoah	020802030601	1660	918

1 Maps were generated to compare the aquatic and terrestrial critical acid loads in each  
2 watershed to determine which estimate provided the greatest protection against acidifying  
3 nitrogen and sulfur deposition. In each watershed, the terrestrial critical load estimate was  
4 compared against each aquatic critical load, and the load with the lowest value was set to  
5 represent the most sensitive component in the watershed. All critical load estimates were  
6 converted to eq/ha/yr for the comparisons.

## 7 8 Results

9 Maps indicating and comparing the sensitivities of the terrestrial and aquatic critical loads  
10 to nitrogen and sulfur deposition in each watershed of the Adirondacks and Shenandoah Case  
11 Study Areas are presented in **Figures 1-4** and **Tables 6-9**.

12  
13 In the Adirondacks Case Study Area, 7 of the 16 watersheds had terrestrial critical acid  
14 loads (based on a Bc/Al of 10.0) that were lower and therefore more sensitive to acidification  
15 than all the lakes in the watershed. However, when the terrestrial critical loads were calculated  
16 with a Bc/Al soil solution ratio of 1.2, only 5 of the 16 watersheds were protected by a terrestrial  
17 critical load that was lower than the aquatic critical loads of the lakes. Three watersheds in the  
18 Adirondacks Case Study Area had terrestrial critical loads (based on a Bc/Al of 10.0) that were  
19 lower and higher than the critical loads for the lakes in the watershed, and one watershed had a  
20 similar mixture of aquatic versus terrestrial acid load protections for terrestrial critical loads  
21 estimated with a Bc/Al of 1.2. In general, a main trend in the Adirondacks Case Study Area was  
22 that watersheds with “Highly Sensitive” and “Moderately Sensitive” lakes were more protected  
23 by aquatic than terrestrial critical acid loads, while the watersheds with “Low Sensitivity” and  
24 “Not Sensitive” lakes were more protected by terrestrial critical acid loads.

25  
26 Similar trends were found in the Shenandoah Case Study Area. However, there was little  
27 distinction between terrestrial acid loads that were calculated with a Bc/Al of 10.0 versus 1.2.  
28 Terrestrial critical acid loads offered a higher level of protection than did the stream aquatic  
29 critical loads in only one watershed. The two lakes in this watershed had “Low Sensitivity” or  
30 were “Not Sensitive” to acidifying nitrogen and sulfur deposition. The 15 watersheds that had  
31 streams with aquatic critical loads lower and more protective than the terrestrial critical loads,

1 were all “Highly Sensitive” or “Moderately Sensitive” to acidifying nitrogen and sulfur  
2 deposition.

3

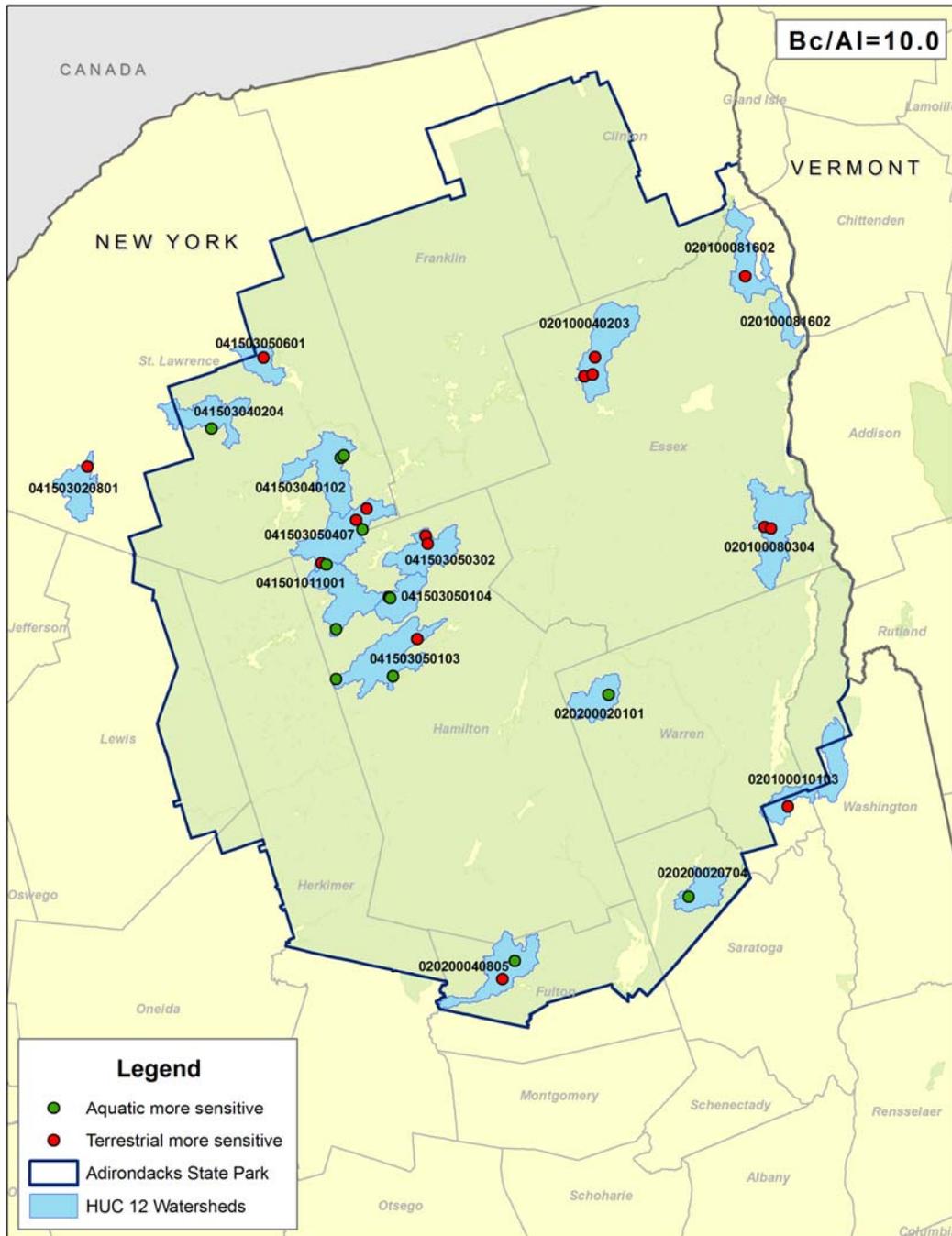
4 In summary, a comparison of the terrestrial and aquatic critical acid loads for watersheds  
5 in the Adirondacks and Shenandoah Case Study Areas indicated that, in general, the aquatic  
6 critical acid loads offered greater protection to the watersheds than did the terrestrial critical  
7 loads. In situations where the terrestrial loads were more protective, the lakes or streams in the  
8 watershed were rated as having “Low Sensitivity” or “Not Sensitive” to acidifying nitrogen and  
9 sulfur deposition. Conversely, when the water bodies were more sensitive to deposition  
10 (“Highly Sensitive” or “Moderately Sensitive”), the aquatic critical acid loads consistently  
11 provided a greater level of protection against acidifying nitrogen and sulfur deposition in the  
12 watershed.

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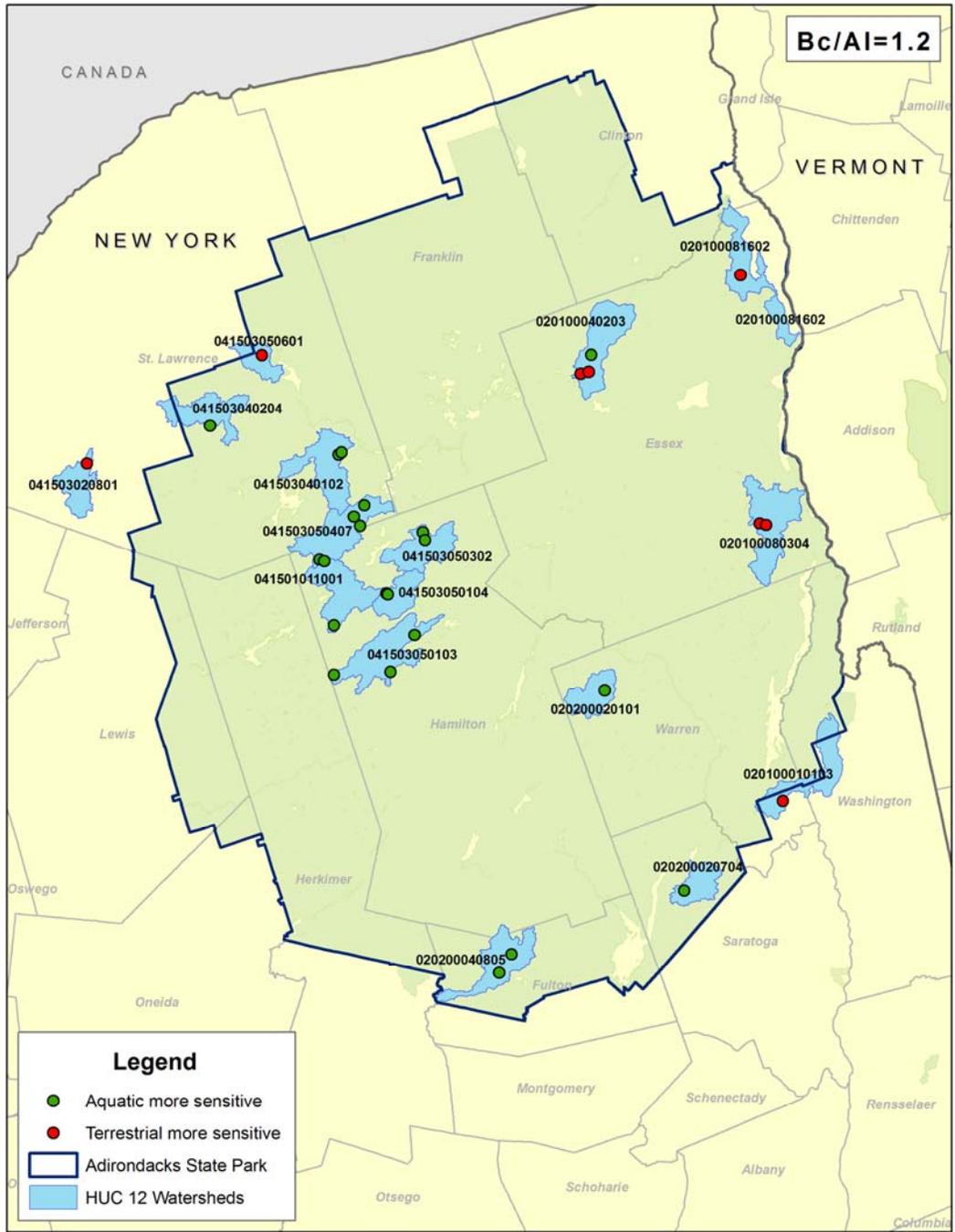
15

1 **Figure 1.** Comparison of aquatic and terrestrial critical loads of acidification (in eq/ha/yr) in the  
 2 16 watersheds of the Adirondacks Case Study Area, based on an ANC of 50 eq/L for the aquatic  
 3 loads and a Bc/AI of 10.0 for the terrestrial loads. Colored circles indicate the locations of the  
 4 water bodies within each watershed. Green circles indicate lakes with critical load values less  
 5 than the terrestrial critical load for the same watershed. Red circles indicate a condition where  
 6 the terrestrial critical load is lower than the lake critical load.  
 7



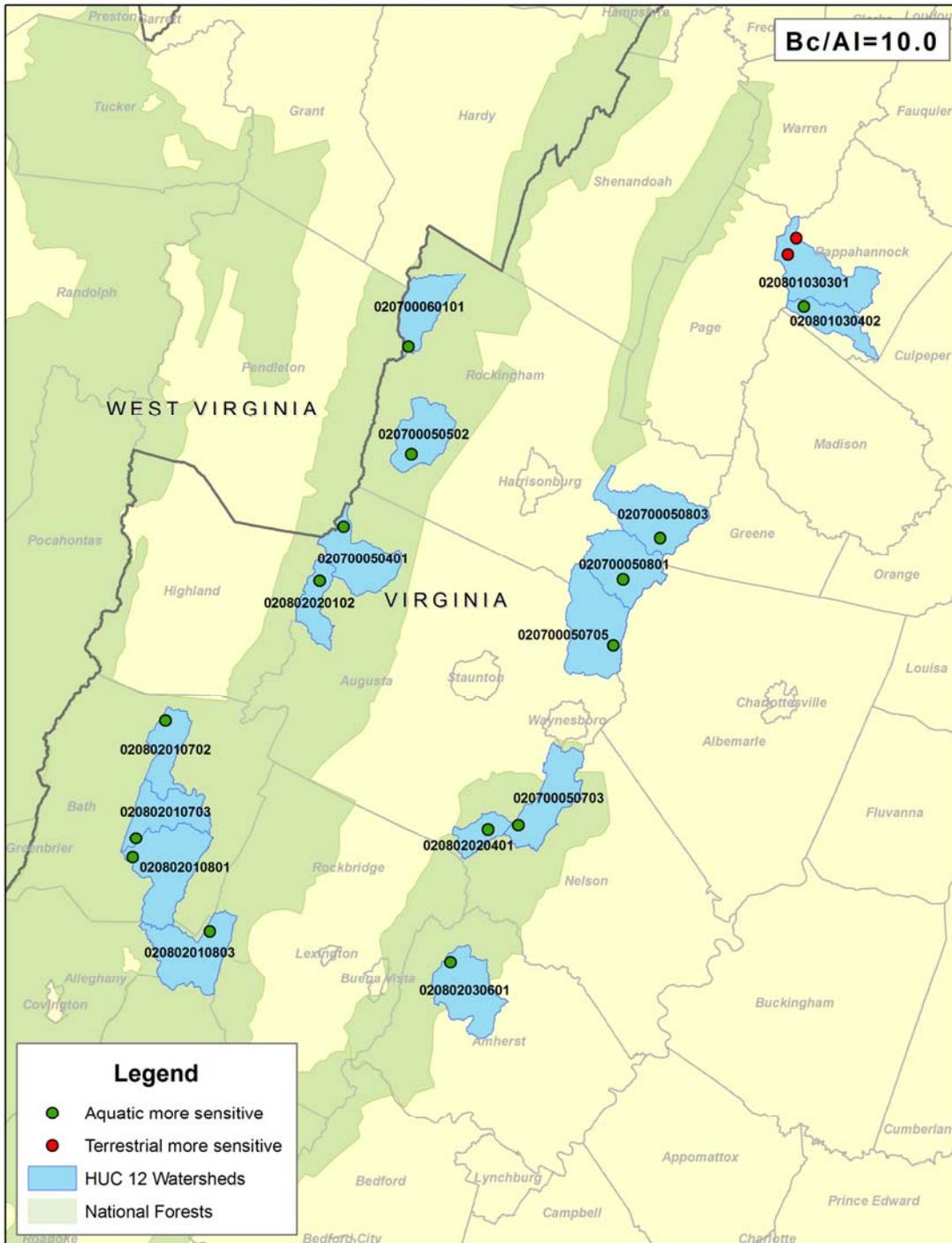
8

1 **Figure 2.** Comparison of aquatic and terrestrial critical loads of acidification (in eq/ha/yr) in the  
 2 16 watersheds of the Adirondacks Case Study Area, based on an ANC of 50 eq/L for the aquatic  
 3 loads and a Bc/AI of 1.2 for the terrestrial loads. Colored circles indicate the locations of the  
 4 water bodies within each watershed. Green circles indicate lakes with critical load values less  
 5 than the terrestrial critical load for the same watershed. Red circles indicate a condition where  
 6 the terrestrial critical load is lower than the lake critical load.  
 7



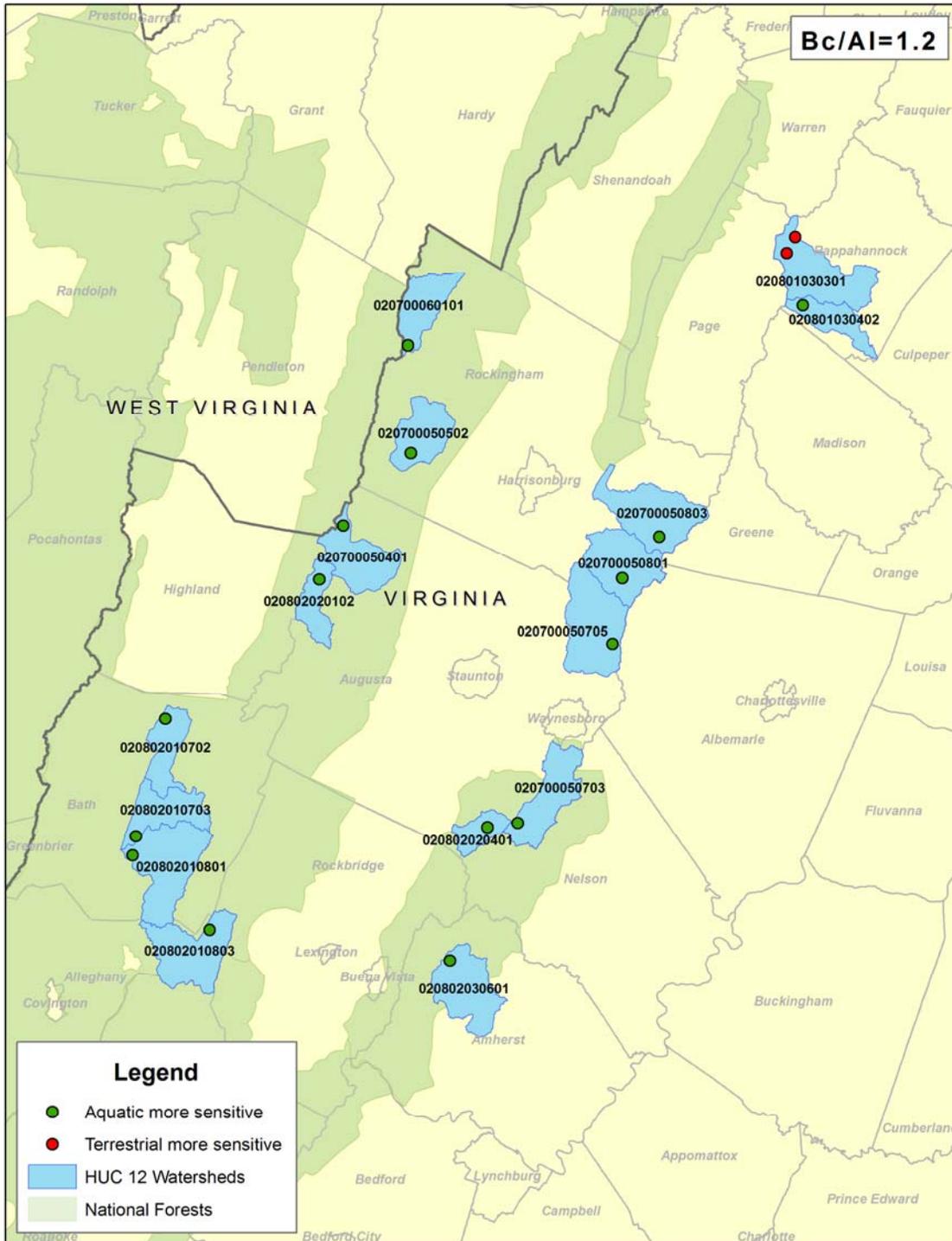
8  
9

1 **Figure 3.** Comparison of aquatic and terrestrial critical loads of acidification (in eq/ha/yr) in the  
 2 16 watersheds of the Shenandoah Case Study Area, based on an ANC of 50 eq/L for the aquatic  
 3 loads and a Bc/AI of 10.0 for the terrestrial loads. Colored circles indicate the locations of the  
 4 waters bodies within each watershed. Green circles indicate streams with critical load values  
 5 less than the terrestrial critical load for the same watershed. Red circles indicate a condition  
 6 where the terrestrial critical load is lower than the stream critical load.  
 7



8

1 **Figure 4.** Comparison of aquatic and terrestrial critical loads of acidification (in eq/ha/yr) in the  
 2 16 watersheds of the Shenandoah Case Study Area, based on an ANC of 50 eq/L for the aquatic  
 3 loads and a Bc/AI of 1.2 for the terrestrial loads. Colored circles indicate the locations of the  
 4 waters bodies within each watershed. Green circles indicate streams with critical load values  
 5 less than the terrestrial critical load for the same watershed. Red circles indicate a condition  
 6 where the terrestrial critical load is lower than the stream critical load.  
 7



8

**Table 6.** Relative sensitivities of aquatic versus terrestrial critical loads in the 29 lakes and 16 watersheds of the Adirondack Case Study Area (based on an ANC of 50 µeq/L for the aquatic loads and a Bc/Al of 10.0 for the terrestrial critical loads and common unit of eq/ha/yr) to acidifying nitrogen and sulfur deposition. Lake IDs are indicated in each cell and are from the REA REPORT (US EPA, 2009). Green text indicates lakes where the aquatic critical load was less than the terrestrial critical load value for the watershed. Red text indicates a condition where the terrestrial critical load for the watershed was lower than the aquatic critical load for the lake within the same watershed.

HUC	CURRENT CONDITION OF ACIDITY AND SENSITIVITY TO ACIDIFICATION CATEGORY			
	Highly Sensitive (CL ≤ 50 meq/m <sup>2</sup> /yr)	Moderately Sensitive (CL = 51-100 meq/m <sup>2</sup> /yr)	Low Sensitivity (CL = 101-200 meq/m <sup>2</sup> /yr)	Not Sensitive (CL > 201 meq/m <sup>2</sup> /yr)
020100010103				NY534L
020100040203			1A2-028O NY310L	NY308L
020100080304				NY312L NY313L
020100081602				NY500L
020200020101		NY013L		
020200020704		NY536L		
020200040805	1A2-078O		NY292L	
041501011001	NY029L			
041503020801				NY783L
041503040102	NY284L NY285L			
041503040204		NY278L		
041503050103	1A1-089O	050215AO NY793L		
041503050104	NY290L			

HUC	CURRENT CONDITION OF ACIDITY AND SENSITIVITY TO ACIDIFICATION CATEGORY			
	Highly Sensitive (CL ≤ 50 meq/m <sup>2</sup> /yr)	Moderately Sensitive (CL = 51-100 meq/m <sup>2</sup> /yr)	Low Sensitivity (CL = 101-200 meq/m <sup>2</sup> /yr)	Not Sensitive (CL > 201 meq/m <sup>2</sup> /yr)
	NY289L			
041503050302			NY008L NY007L	
041503050407		NY767L NY529L NY528L NY769L	NY768L	
041503050601			NY004L	

**Table 7.** Relative sensitivities of aquatic versus terrestrial critical loads in the 29 lakes and 16 watersheds of the Adirondack Case Study Area (based on an ANC of 50 µeq/L for the aquatic loads and a Bc/Al of 1.2 for the terrestrial critical loads and common unit of eq/ha/yr) to acidifying nitrogen and sulfur deposition. Lake IDs are indicated in each cell and are from the REA REPORT (US EPA, 2009). Green text indicates lakes where the aquatic critical load was less than the terrestrial critical load value for the watershed. Red text indicates a condition where the terrestrial critical load for the watershed was lower than the aquatic critical load for the lake within the same watershed.

HUC	CURRENT CONDITION OF ACIDITY AND SENSITIVITY TO ACIDIFICATION CATEGORY			
	Highly Sensitive (CL ≤ 50 meq/m <sup>2</sup> /yr)	Moderately Sensitive (CL = 51-100 meq/m <sup>2</sup> /yr)	Low Sensitivity (CL = 101-200 meq/m <sup>2</sup> /yr)	Not Sensitive (CL > 201 meq/m <sup>2</sup> /yr)
020100010103				NY534L
020100040203			1A2-028O NY310L	NY308L
020100080304				NY312L NY313L
020100081602				NY500L
020200020101		NY013L		
020200020704		NY536L		
020200040805	1A2-078O		NY292L	
041501011001	NY029L			
041503020801				NY783L
041503040102	NY284L NY285L			
041503040204		NY278L		
041503050103	1A1-089O	050215AO NY793L		

HUC	CURRENT CONDITION OF ACIDITY AND SENSITIVITY TO ACIDIFICATION CATEGORY			
	Highly Sensitive (CL ≤ 50 meq/m <sup>2</sup> /yr)	Moderately Sensitive (CL = 51-100 meq/m <sup>2</sup> /yr)	Low Sensitivity (CL = 101-200 meq/m <sup>2</sup> /yr)	Not Sensitive (CL > 201 meq/m <sup>2</sup> /yr)
041503050104	NY290L NY289L			
041503050302			NY008L NY007L	
041503050407		NY767L NY529L NY528L NY769L	NY768L	
041503050601			NY004L	

**Table 8.** Relative sensitivities of aquatic versus terrestrial critical loads in the 20 streams and 16 watersheds of the Shenandoah Case Study Area (based on an ANC of 50 µeq/L for the aquatic loads and a Bc/Al of 10.0 for the terrestrial critical loads and common unit of eq/ha/yr) to acidifying nitrogen and sulfur deposition. Stream IDs are indicated in each cell and are from the REA REPORT (US EPA, 2009). Green text indicates streams where the aquatic critical load was less than the terrestrial critical load value for the watershed. Red text indicates a condition where the terrestrial critical load for the watershed was lower than the aquatic critical load for the stream within the same watershed.

HUC	CURRENT CONDITION OF ACIDITY AND SENSITIVITY TO ACIDIFICATION CATEGORY			
	Highly Sensitive (CL ≤ 50 meq/m <sup>2</sup> /yr)	Moderately Sensitive (CL = 51-100 meq/m <sup>2</sup> /yr)	Low Sensitivity (CL = 101-200 meq/m <sup>2</sup> /yr)	Not Sensitive (CL > 201 meq/m <sup>2</sup> /yr)
020700050401	VT37			
020700050502	VT57			
020700050703	VT40			
020700050705	VT35 VT36			
020700050801	DR01 WOR1			
020700050803	VT53			
020700060101		VT54		
020801030301			VT60	VT61
020801030402		VT62		
020802010702	VT10			
020802010703	VT11	VT12		
020802010801	VT14 VT15			

HUC	CURRENT CONDITION OF ACIDITY AND SENSITIVITY TO ACIDIFICATION CATEGORY			
	Highly Sensitive (CL ≤ 50 meq/m <sup>2</sup> /yr)	Moderately Sensitive (CL = 51-100 meq/m <sup>2</sup> /yr)	Low Sensitivity (CL = 101-200 meq/m <sup>2</sup> /yr)	Not Sensitive (CL > 201 meq/m <sup>2</sup> /yr)
020802010803	VT16			
020802020102		VT38		
020802020401	VT41			
020802030601		VT46		

**Table 9.** Relative sensitivities of aquatic versus terrestrial critical loads in the 20 streams and 16 watersheds of the Shenandoah Case Study Area (based on an ANC of 50 µeq/L for the aquatic loads and a Bc/Al of 1.2 for the terrestrial critical loads and common unit of eq/ha/yr) to acidifying nitrogen and sulfur deposition. Stream IDs are indicated in each cell and are from the REA REPORT (US EPA, 2009). Green text indicates streams where aquatic critical loads were less than the terrestrial critical load value for the watershed. Red text indicates a condition where the terrestrial critical load for the watershed was lower than the aquatic critical load for the stream within the same watershed.

HUC	CURRENT CONDITION OF ACIDITY AND SENSITIVITY TO ACIDIFICATION CATEGORY			
	Highly Sensitive (CL ≤ 50 meq/m <sup>2</sup> /yr)	Moderately Sensitive (CL = 51-100 meq/m <sup>2</sup> /yr)	Low Sensitivity (CL = 101-200 meq/m <sup>2</sup> /yr)	Not Sensitive (CL > 201 meq/m <sup>2</sup> /yr)
020700050401	VT37			
020700050502	VT57			
020700050703	VT40			
020700050705	VT35 VT36			
020700050801	DR01 WOR1			
020700050803	VT53			
020700060101		VT54		
020801030301			VT60	VT61
020801030402		VT62		
020802010702	VT10			
020802010703	VT11	VT12		
020802010801	VT14 VT15			

HUC	CURRENT CONDITION OF ACIDITY AND SENSITIVITY TO ACIDIFICATION CATEGORY			
	Highly Sensitive (CL ≤ 50 meq/m <sup>2</sup> /yr)	Moderately Sensitive (CL = 51-100 meq/m <sup>2</sup> /yr)	Low Sensitivity (CL = 101-200 meq/m <sup>2</sup> /yr)	Not Sensitive (CL > 201 meq/m <sup>2</sup> /yr)
020802010803	VT16			
020802020102		VT38		
020802020401	VT41			
020802030601		VT46		

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1

2

### APPENDIX C: Elasticity AAPI sensitivity analyses

3

**Elasticity.** One metric for determining sensitivity of the AAPI to its component parameters is elasticity. Elasticity measures the percent change in AAPI for a one percent change in the component. In general, the formula for an elasticity is:

6

$$E_{X_j}^{AAPI} = \frac{\partial AAPI}{\partial X_j} \cdot \frac{X_j}{AAPI}$$

8

9 Where  $E_{X_j}^{AAPI}$  is the elasticity of AAPI with respect to component  $X_j$ , and  $j$  is the number of components.

10

11 So, for AAPI defined as

12

$$AAPI = \frac{1}{Q} N_{eco} + BC_0^* - \frac{1}{Q} NHx - \frac{1}{Q} [T_{NOy} \cdot NOy + T_{SOx} \cdot SOx]$$

14

15 The set of relevant elasticities are:

16

17 For runoff, Q:

18

$$E_Q^{AAPI} = -\frac{1}{Q^2} [N_{eco} - NHx - T_{NOy} NOy - T_{SOx} SOx] \times \frac{Q}{AAPI}, \text{ which can be rewritten as}$$

20

$$E_Q^{AAPI} = -\frac{1}{Q} [AAPI - BC_0^*] \times \frac{Q}{AAPI}, \text{ or}$$

22

1  $E_Q^{AAPI} = -1 + \frac{BC_0^*}{AAPI}$

2

3 For  $BC_0^*$ ,

4

5  $E_Q^{AAPI} = \frac{BC_0^*}{AAPI}$

6

7 For  $Neco$ ,

8

9  $E_{Neco}^{AAPI} = \frac{1}{Q} \cdot \frac{Neco}{AAPI}$

10

11 For  $NHx$ ,

12

13  $E_{NHx}^{AAPI} = -\frac{1}{Q} \cdot \frac{NHx}{AAPI}$

14

15 For  $T_{NOy}$ ,

16

17  $E_{T_{NOy}}^{AAPI} = -\frac{1}{Q} \cdot NOy \cdot \frac{T_{NOy}}{AAPI}$

18

19 For  $T_{SOx}$ ,

20

21

1 
$$E_{T_{SOx}}^{AAPI} = -\frac{1}{Q} \cdot SOx \cdot \frac{T_{SOx}}{AAPI}$$

2

3 For NOy,

4

5 
$$E_{NOy}^{AAPI} = -\frac{1}{Q} \cdot T_{NOy} \cdot \frac{NOy}{AAPI}$$

6

7 For SOx,

8

9 
$$E_{SOx}^{AAPI} = -\frac{1}{Q} \cdot T_{SOx} \cdot \frac{SOx}{AAPI}$$

10

11 These elasticities can be evaluated at various points along the ranges of each component, as well as along  
12 ranges of the AAPI. We evaluate the elasticities at the sample means, medians, first quartiles, and third  
13 quartiles. Elasticities are evaluated only for ecoregions that overlap the CMAQ modeling domain which  
14 provides values for reduced nitrogen and the transformation ratios ( $T_{NOx}$  and  $T_{SOx}$ ). This will provide a  
15 reasonable assessment of the sensitivity of the AAPI to input components. Table 1 provides the estimated  
16 elasticities. Elasticities are summarized across ecoregions using means, medians, minimums, and  
17 maximums.

18 Note that elasticities can be either positive or negative. A negative elasticity means that the calculated  
19 AAPI will decrease as a component increases. The magnitude of the elasticity depends on the values of  
20 the components and the starting value of AAPI.

21 Based on the calculated elasticities, AAPI is most responsive to changes in Q, BC0, and Neco with some  
22 responsiveness to reduced N. Note that for some components, such as Q, the elasticities switch signs  
23 depending on the values of the variables for which the elasticity is evaluated. This suggests potentially  
24 important interactions. AAPI is not responsive to the transformation ratios, TNOx and TSOx at mean  
25 values of the AAPI components. However, when the elasticities for TNOx and TSOx are evaluated at the  
26 first quartiles of the data, some locations in the Eastern U.S. show higher responsiveness to changes in  
27 TNOx and TSOx, with elasticities as high as 2.

28

29

1 **Table 1. Elasticity\* of AAPI to Component Variables**

<b>AAPI Component</b>	<b>Metric for Which Elasticities are Evaluated</b>	<b>Mean Elasticity Across Ecoregions</b>	<b>Median Elasticity Across Ecoregions</b>	<b>Minimum Elasticity Across Ecoregions</b>	<b>Maximum Elasticity Across Ecoregions</b>
Runoff (Q)	Mean	-0.1047	0.1221	-20.4572	1.6005
	Median	0.2561	0.1426	-6.2481	2.4578
	1 <sup>st</sup> Quartile	-0.9283	0.1303	-135.2544	88.1063
	3 <sup>rd</sup> Quartile	0.2988	0.1110	-0.1810	7.5684
Base Cation Weathering (BC0)	Mean	0.8953	1.1221	-19.4572	2.6005
	Median	1.2561	1.1426	-5.2481	3.4578
	1 <sup>st</sup> Quartile	0.0717	1.1303	-134.2544	89.1063
	3 <sup>rd</sup> Quartile	1.2988	1.1110	0.8190	8.5684
Neco	Mean	0.0179	0.1464	-13.8545	1.6051
	Median	0.3376	0.2563	-4.7203	2.5044
	1 <sup>st</sup> Quartile	0.3543	0.2596	-115.1112	137.0526
	3 <sup>rd</sup> Quartile	0.3016	0.1440	0.0137	6.5565
Reduced Nitrogen	Mean	-0.0409	-0.0702	-0.4708	3.9332
	Median	-0.1407	-0.1031	-0.7957	1.0061
	1 <sup>st</sup> Quartile	-0.0308	-0.1190	-33.9063	35.3783
	3 <sup>rd</sup> Quartile	-0.1128	-0.0615	-1.9167	-0.0050
NOx Transformation Ratio (T <sub>NOx</sub> )	Mean	0.0089	-0.0053	-0.0597	1.0598
	Median	-0.0061	-0.0064	-0.0506	0.2751
	1 <sup>st</sup> Quartile	-0.0191	-0.0071	-2.7061	1.7749
	3 <sup>rd</sup> Quartile	-0.0154	-0.0044	-0.5028	-0.0002

<b>AAPI Component</b>	<b>Metric for Which Elasticities are Evaluated</b>	<b>Mean Elasticity Across Ecoregions</b>	<b>Median Elasticity Across Ecoregions</b>	<b>Minimum Elasticity Across Ecoregions</b>	<b>Maximum Elasticity Across Ecoregions</b>
SOx Transformation Ratio ( $T_{SOx}$ )	Mean	0.0019	-0.0024	-0.0185	0.3608
	Median	-0.0045	-0.0036	-0.0413	0.1091
	1 <sup>st</sup> Quartile	0.0040	-0.0032	-1.8023	1.9860
	3 <sup>rd</sup> Quartile	-0.0058	-0.0021	-0.1534	-0.0002

1

2 \*Elasticity is the percent change in AAPI for a one percent change in the component variable.  
3 For example, when evaluated at the means of all component variables, the mean elasticity of  
4 AAPI to the runoff variable Q is -0.1047, which means that for each 1 percent increase in Q, the  
5 AAPI is reduced by 0.1047 percent.

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