Comments on

RISK AND EXPOSURE ASSESSMENT FOR REVIEW OF THE SECONDARY NATIONAL AMBIENT AIR QUALITY STANDARDS FOR OXIDES OF NITROGENAND OXIDES OF SULFUR

SECOND DRAFT

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These comments address several key scientific issues related to the applicability of using regional ecosystem modeling based on ecological indicators and deposition thresholds to assign secondary NOx and SOx national ambient air quality standards (NAAQS) for welfare effects.

Two main issues stand out from the EPA Risk and Exposure Assessment (REA):

1. Model assumptions used in the case study analysis are not applicable to the entire case study region let alone the entire United States.
2. Other environmental factors, including spatial differences, exist across the case study regions as well as the United States that determine how an ecosystem responds to atmospheric deposition loading of nitrogen and sulfur.

These key concerns are discussed below, followed by detailed comments on the specific sections of the REA covering aquatic/terrestrial acidification and aquatic/terrestrial nutrient enrichment.

Model assumptions are not applicable to the entire case study region let alone the entire United States

There are several modeling assumptions employed in the REA that are not adequate to determine biological levels of protection within the case study regions let alone over the entire United States. These include the assumption of steady-state and model assumptions that are too simple to adequately capture complex processes. In addition, there is the hurdle of separating atmospheric deposition effects from "background" or "pre-acidification" conditions.

Several of the models (SSWC, SMB, and SPARROW) assume steady-state and mass balance conditions. However the steady-state nature of the models is not adequately represented. For example, both the SSWC model and the SMB assume that base cations in a catchment are equal to the acid anions entering the system. However, the literature also suggests that the deposition of acid anions themselves entering the system can increase base cation leaching through ion-exchangers in the soil. The SSWC model attempts to account for this by using the f-factor to account for pre-industrial acidification; however, lack of research for the U.S. regions makes it inappropriate to apply default values from European countries across the U.S. with any degree of confidence. In fact, the Shenandoah case study used an entirely different approach to hindcast the base cation supply to 1860 because of the lack of confidence in using the f-factor in an area that had not been previously tested with this approach (EPA, 2009). This factor can vary spatially and is dependent upon site specific catchment features and lake chemistry. Within the European approach for calculating f-factor, a range of values are suggested. However, the REA uses a

single default value based on empirical studies from Europe (see Appendix 4, Attachment A-13) rather than a range to explore uncertainty. The following questions should have been addressed in the REA:

- How to account for the spatial distribution of base cation supply and the f-factor if the case study is broadly applied across the US?
- Can the case study verify through empirical studies that 400 μeq/L is the correct assumption for the region and that this assumption can be extended to other region of the United States?

In addition to spatial heterogeneity, temporal variation in the f-factor may also be of concern when using a steady-state model. Lake chemistry determines the f- factor; however, year-to-year variations cannot be taken into effect in the steady-state model, and Watmough et al. (2005) demonstrates this by showing how significant variances in lake chemistry over a 13-year period (collected in 1985 and 1998) result in different f-factors when derived from any one year.

The base cation uptake variable used in both SMB and SSWC models assumes that biomass harvesting is the only permanent sink for base cation retention and uptake, and when handled inconsistently could influence the critical load levels. Watmough and Dillon (2002) compared how base cation flux was modeled in both the SMB and the SSWC models. They determined that, when the SSWC model did not account for base cation uptake due to harvesting, critical load values were higher compared to model-runs that assumed harvesting. These issues raise some important questions that should have been addressed in the REA:

- How will the EPA implement the ecological effect function across the entire United States when not all ecosystems are as well understood as the case studies?
- When extending critical loads modeling to a broader area using the Forest Inventory and Analysis National Program (FIA) data, how was the removal of biomass across the US or in larger study areas generalized?

In the two case studies for the REA, denitrification was considered negligible because the regions are upland forests; therefore the value was set to 0 eq/ha/yr. However when the case study areas were expanded the REA did not provide a method for calculating denitrification and used the case study value of no denitrification. This leads to the question:

- How will the EPA determine this input value across other ecosystems in the U.S. where denitrification processes may be important but the rate of denitrification is not known?

The SMB model does not take into account any increased net uptake of nitrogen through aggrading stands because of the steady-state assumption. Additional problems with assuming steady-state are the temporal limitations that cannot account for chronic versus acute acidity and seasonality; seasonality influences the vegetation (including submerged aquatic vegetation) and temperature that can in turn increase mineralization processes. Temporal components such as lag time for ecosystem response or relationships between stages of nutrient enrichment and deposition are not modeled. The aquatic nutrient enrichment case study uses SPARROW, a statistical/linear process model as a mass balance; however, this methodology is highly uncertain (as pointed out in the REA) in trying to predict processes in nature that could respond in a non-linear fashion, such as fitting a logistic response when back-calculating the atmospheric NOx load reductions needed to improve estuary conditions.

Other model assumptions make it difficult to apply the case study areas to other parts of the United States. This includes assuming a default value for root zone depth (in determining base cation weathering), treating soil as one homogeneous layer, and ignoring nutrient cycling between plants and soils.

- In two of the case studies, only one ecological indicator (Bc:Al or ANC) is used to represent biological response. There is no justification provided in the REA if one indicator is adequate or the most representative.

There is also a lack of standardized methodology for determining model input parameters. The REA used the clay-substrate method and the soil-type texture method to determine base cation weathering. However several other methods exist in the literature for use. These include assigning a Skokloster class for soil type, base mineral index correlation, total cation content correlation, and use of the model PROFILE. The uncertainty about which value to use is shown in the REA by using two methods for calculating weathering rate, two gibbsite equilibrium values from the literature (300 and 3,000 m^6 eq^2), and two methods to account for base cation and nitrogen uptake (harvesting vs. no harvesting). This resulted in various critical load values as shown in the Appendix 5, Tables 3.1-1 through 3.1-8. Several studies have shown that when using an incorrect gibbsite factor or not accounting for the relationship between Al^{3+} and H^+ in soil solution, the critical load for acidity can be skewed higher or lower (Holmberg et al., 2001). The different gibbsite values are site specific and are determined by the percent organic matter in the soil (UBA, Mapping Manual, 1996). Altering this variable over a range of recommended values for different soil types according to a European Mapping Manual changed UK critical loads in one study from a mean of 3.2 to 1.8 keq/ha/yr (Skeffington, 2006; Holmberg et al, 2001).

- The REA does not explain how to assign an appropriate critical load value when such a wide range can be calculated depending on the assumptions used for these two case studies.

The REA does not discuss how lowering one reactive form of nitrogen (NOx) results in a concomitant linear decrease of total nitrogen deposition. This also extends to the limitation of using the SMB to evaluate alternative depositions on the biological level of protection, because the REA does not differentiate between different reactive nitrogen species. The case study for aquatic nutrient enrichment fails to explain the linkage between SPARROW and ASSETS EI modeling and appears to be incomplete at the time of the REA second draft. This is pointed out in the need for future model parameterization concerning calibration timeframes and deposition data. The ASSETS EI approach assumes that the system in not naturally eutrophic, another assumption that may not extend to different parts of the United States. Not only are the aquatic nutrient enrichment case study response curves theoretically-based, but they are also based on expert judgment for the determined future outlook index value for watershed management options.

In summary, these assumptions are too simple to account for the multitude of factors that affect cycling of substances in terrestrial and aquatic ecosystems.

Other environmental factors and spatial differences exist across the case study region as well as the United States that determine how ecosystems respond to atmospheric deposition of N and S

A multitude of factors influence nitrogen retention in terrestrial ecosystems over different spatial and temporal scales. Temperature influences mineralization rates, thus impacting storage (Fenn et al., 1998; Mitchell et al., 1996). Nitrate leaching is generally greater in mature forests as opposed to younger forests, as nutrient incorporation declines as the system approaches steady-state (Vitousek and Reiners, 1975; Barker et al., 2002; Goodale et al., 2000). Prior land-use history and the extent to which nitrogen loss via agricultural clearing, forest fires (Fenn et al., 2003), and timber harvesting (Pardo et al., 1995; Goodale et al., 2002) can resemble earlier stages of stand development and drive ecosystems back to a state of higher nitrogen demand (Aber et al., 1998; Johnson and Lindberg, 1992). There are several issues as to why one biological level of protection set for one case study is not generalizable to the US as a whole and the REA has not attempted to address those issues.

Across the U.S., a variety of physical, biological, meteorological, and climate factors exist. Each of these parameters in turn influences how a water body or ecosystem may receive or respond to deposition of nitrogen and sulfur from both the standpoint of acidity and nutrient enrichment. As each of these influencing processes is highly variable, so is the ecosystem response. The REA attempts to quantify the

risk associated with atmospheric deposition back to ecological harm; however, it is difficult to model or predict natural occurrences that are spontaneous, extreme, or driven by uncontrollable factors. While the REA case studies use simple ecosystem modeling in an attempt to represent the response to both nitrogen and sulfur deposition, the REA does not address occurrences such as fire, windstorms, insect invasion, and historic land use that may have unknowingly altered the ecosystem; drought; or groundwater sources that may influence nitrogen cycling.

- Although the case studies examine risk for acidification or nutrient enrichment, their results yielded great uncertainty. Across the entire United States some areas are more acid-sensitive because of acidifying compounds that exist under natural conditions or some areas will not respond at all. The REA does not address how these factors will be overcome to determine a uniform standard.

The difficulty of determining a uniform standard is highlighted by the existence of a variety of geology across the U.S. and the challenge in characterizing the weathering process for determining the response to acidification in both aquatic and nutrient ecosystems. The case studies evaluated in the REA are site specific for soil types, bedrock geology, age of systems, and existing data that may be able to identify and point out natural causes of acidity. However, the REA does not adequately apply this concept across the entire United States.

As an example, it was acknowledged by many scientists in the 1970s and 1980s that many of the lakes present in the upper Midwest (states of MN, WI, and MI) represented a unique challenge to air quality regulators who were justifiably concerned about acid deposition impacts to these resources (Eilers et al., 1988). The increase in SO_2 emissions domestically between the early 1900s and the 1980s and the sulfate deposition attributed to these emissions had been associated with aquatic acidification in the Northeast United States, Canada and as far as Scandinavia. It soon became apparent that water chemistry and watershed factors were important parameters for determining the sensitivity of Wisconsin lakes to acidification. Hydrological factors (e.g., the degree to which the watershed interacted with subject lakes) allowed one to predict susceptibility to acidification (Eilers et al., 1983). However, even among Wisconsin lakes that received much if not most of their water from precipitation there was surprising resistance to ambient and modeled increases in deposition due to small but significant inputs from ground water (Garrison et al., 1987; Webster et al., 1993).

The early research and water quality monitoring identified the potential susceptibility of lakes in the upper Midwest to acidification. Yet, in the context of the 1980s, and in spite of wet sulfate deposition that was occurring in this region, comparisons between historic (1921-1941) and more contemporary (1979-1983)

water quality measurements (Eilers et al., 1989) showed that the vast majority of the 149 lakes, for which data were available, experienced no change or even slight increases in ANC; only a few lakes showed slight declines. Interestingly, by the mid-1980s, regional experts in the topic of lake acidification could not rule out natural sources of acidification (e.g., wetland/bog encroachment, presence of mineral soils) as a cause for the low ANC chemical conditions observed in this region (Weiner and Eilers, 1987).

Based on NADP wet deposition monitoring since the late 1970s in the Upper Midwest (NADP, 2008), acid and sulfate deposition has since declined significantly from levels that were, in the context of the early 1980s, believed by some researchers to be injurious to lakes in this region based on data from the Northeastern United States and Scandinavia. The occurrence of drought conditions in this area has complicated simplistic comparisons between changes in lake water and precipitation chemistry. For example, some benchmark lakes have exhibited reductions in sulfate but ANC levels have dropped due to lower inputs of alkaline groundwater caused by the drought (Webster and Brezonik, 1995).

The case study selection process in the EPA Integrated Science Assessment and the REA focused on areas that were plentiful, although still not completely sufficient, in data resources as a result of long-term research of acidic deposition. However, the existing scale of national databases required to extend ecosystem modeling across the U.S. to support a NAAQS do not exist for several key parameters. This is especially true with regards to reduced forms of nitrogen which are not currently monitored on a standardized basis across the United States. Default values were used for important model inputs (such as with denitrification and nitrogen immobilization in the terrestrial acidification case study) to determine site specific critical loads; while expert judgment was used in other case studies (ASSETS EI in the aquatic nutrient enrichment case study).

- The reliance on model default values or expert judgment in the REA to model ecosystems highlights the need for future research and monitoring in order to adequately evaluate the relationship between atmospheric deposition of total nitrogen and sulfur and the response of ecological indicators to ambient air concentrations.

One of the most important issues of concern is data availability. Three important model input parameters (base cation weathering, base cation uptake, and ANC based on Bc:Al) are site specific and are estimated in the REA. Default values from a literature review (McNulty et al., 2007; ICP, 2004) were used for denitrification, nitrogen immobilization, rooting zone soil depth, and the gibbsite equilibrium. The EPA recognizes the importance of estimating accurate weathering rates as input parameters in SMB modeling and has used two methods to determine soil weathering rate in the absence of actual data for case study areas. The soil weathering rates were then corrected for soil temperature and rooting zone depth. The

REA then extended the critical load process into additional ecosystem ranges where the selected indicator species exist in order to generalize the linkage between deposition and critical loads modeling. However, calculations of critical loads to the expanded areas were not held to as high a standard as in the case studies. This is especially true with regard to estimating base cation weathering because of the lack of data pertaining to parent material and acidity. To estimate acidity and weathering rate for the expanded area, Soil Survey Geographic Database (SSURGO) data were used (where available) and the parent material acidity was inferred. In some instances, these data were estimated based on the probable range of soil mineral content. If these data were unavailable, state-level data were used. Base cation weathering was not corrected for temperature in this expanded process.

To account for model uncertainty based on the estimated values of base cation weathering rate, the REA ran uncertainty analysis on the range of potential miscalculation of parent material. Comparing the resulting critical loads revealed that changes in critical load values could range from 0 to 3,631 eq/ha/yr for sugar maple and 0 to 1,584 eq/ha/yr for red spruce, corresponding to percentage differences ranging from 0% to 492% and 0% to 453% for sugar maple and red spruce, respectively (EPA, 2009). This vast range of uncertainty thereby confounds the process for assigning a value that can be then used to determine a uniform national standard.

The comments presented above describe why the proposed REA methodology used to quantify linkages between deposition and ecological effects over a national scale has not been shown to be adequate for translating back to a uniform national standard.

Specific Comments on Aquatic Acidification:

In addition to the comments provided in the main section, we observe the following in the sections of the REA addressing aquatic acidification:

The REA places special emphasis on what is termed the "pre-acidification" concentration of streamwater chemistry and the exceedance of current concentrations. The assessment attempts to relate the exceedances back to a deposition of N and S to define a link between deposition and improved (or maintaining ANC). However, SOx/NOx deposition reductions have often resulted in little improvement in concentration. The REA implies that because streamwater NO_3^- and SO_4^{2-} concentrations are above "pre-acidification" levels modeled from the 1860s, that deposition is the only process impacting surface water chemistry.

- The linkage between streamwater chemistry and deposition is not thoroughly explained in the results section. In addition, MAGIC hindcasts were run on streamwater chemistry to determine

anthropogenic contribution of deposition, but the processes of uptake, loss, and mineralization in the catchment were not explained in the text.

The SSWC model assumes steady-state conditions and for the net uptake variable it is assumed that the only permanent sink is when biomass is harvested from the catchment (EPA, 2009). For the Adirondack Mountains used as the case study for acidification, net uptake is considered zero. However, this is not always the case for ecosystems across the U.S. where harvesting has occurred or could occur at a later date. Watmough and Dillon (2002) compared how BC flux was modeled in both the simple mass balance (SMB) model and the SSWC model. They determined that, when the SSWC model did not account for BC uptake due to harvesting, critical load values were higher compared to model-runs that assumed harvesting.

- EPA has not shown in the REA how it will handle sensitive regions in the US that harvest biomass.

Specific Comments on Terrestrial Acidification:

In addition to the comments provided in the main section, we observe the following in the sections of the REA addressing terrestrial acidification:

A limitation of the SMB model is that while it allows for the analysis of both nitrogen and sulfur deposition loads, it does not allow for the analysis of effects between the different reactive nitrogen species.

- EPA should apply models that take into account the distinct cycling and impacts of different reactive nitrogen species.

For the REA, five of these parameters (BC_{dep}, Cl_{dep}, N_i and N_{de}) use selected and defined input values (EPA, 2009); however three parameters (BC_w, Bc_u and $ANC_{le,\ crit}$) required calculations that are site specific. The REA recognizes that weathering rate is one of the most influential and difficult-to-estimate parameters when modeling critical loads (EPA, 2009). BC weathering refers to the release of base cations from mineral complexes in the soil due to the process of chemical dissolution over time, thereby producing alkalinity and increasing neutralization (ICP, 2004). Two methods, the clay-substrate method (Ouimet et al., 2006; Watmough et al., 2006; McNulty et al., 2007; Pardo and Duarte, 2007) and the soil-type texture method (ICP, 2004), were used to calculate the BC weathering rate in the REA. Additional methods for estimating weathering rate that were not used in the REA include Skokloster classes (ICP, 2004), base mineral index correlation, total cation content correlation

"zirconium method", or the use of PROFILE (a multi-layer hydrogeochemical model, Nilson and Grennflelt, 1988; Hall et al., 2001).

- The REA should have shown results of sensitivity studies using all the methods described above in order to address different weathering rate assumptions and provide guidance as to how to address the variance in results.

Specific Comments on Aquatic Nutrient Enrichment:

In addition to the comments provided in the main section, we observe the following in the sections of the REA addressing aquatic nutrient enrichment:

SPARROW is a combination statistical/process-based model, where the dependent variable (contaminant transport over time) is linearly related to all other sources of contaminant mass in the model (EPA, 2009). One of the main factors of uncertainty in using the SPARROW model to evaluate atmospheric nitrogen loading is that it applies a linear process model to predict the watershed response. The creation of response curves linking SPARROW to the ASSETS EI also assumes a logistic curve that may not exist it nature. The response curves are then used to back calculate atmospheric NO_x load reductions in an attempt to improve estuary conditions. The manner in which the SPARROW regression model handles normal distribution in the system, by applying non-parametric methods, can also lead to an inaccurate representation of actual processes. Currently, expert judgment is used to determine the upper/lower bounding limits for response curve uncertainty. There is uncertainty in drawing conclusions for response based on presence/absence of symptoms of eutrophication that could be influenced by a multitude of factors other than atmospheric deposition. In addition, the REA points out that there is a need for final model parameterization of SPARROW with more current data, and difficulty in expressing land-to-water delivery fractions related to the proportion/fraction of total nitrogen input. The REA discusses the difficulty in understanding the processes leading to eutrophication in estuaries, highlighting that a causal relationship between atmospheric deposition and ecological harm are not fully understood at this time.

- EPA has not addressed the limitation of the SPARROW model in not being able to accurately represent non-linear processes.

- Given the uncertainties in establishing a causal relationship between atmospheric and ecological harm, the uncertainties of the contribution of other factors leading to euthrophication, and the lack of model parameterization with additional data, EPA needs to develop or employ a more robust methodology for performing and evaluating such analyses.

Specific Comments on Terrestrial Nutrient Enrichment:

In addition to the comments provided in the main section, we observe the following in the sections of the REA addressing terrestrial nutrient enrichment:

> The terrestrial nutrient enrichment case study sought to understand the relationship between nitrogen loads and the impact of fire frequency and invasive plants. This was considered because fire and increased nitrogen are thought to enhance non-native grasses in the CSS ecosystem (EPA, 2009). Land cover change data from 1993-1997 (California Department of Forestry and Fire Protection, FRAP data) was used to quantify changes in the amount of CSS loss and fire threat through GIS data interpretation. FRAP data from 2002 was also used to quantify the change in MCF ecosystem (EPA, 2009).
>
> However, the GIS modeling of fire threat did not really demonstrate (but rather speculated) on the influence of nitrogen dynamics on fire. The REA did not show correlations, or try to model any loss (or gain) of fire-dependent species.
>
> - A more robust analysis is needed before definitive conclusions on the relationship between terrestrial nutrient enrichment and fire frequency can be made.

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