



Clean Air Scientific Advisory Committee (CASAC)

A Federal Advisory Committee to the U.S. Environmental Protection Agency

September 27, 2023

EPA-CASAC-23-005

The Honorable Michael S. Regan
Administrator
U.S. Environmental Protection Agency
1200 Pennsylvania Avenue, N.W.
Washington, D.C. 20460

Subject: CASAC Review of the EPA's *Policy Assessment for the Review of the Secondary National Ambient Air Quality Standards for Oxides of Nitrogen, Oxides of Sulfur and Particulate Matter (External Review Draft – May 2023)*

Dear Administrator Regan:

The Clean Air Scientific Advisory Committee (CASAC) Oxides of Nitrogen, Oxides of Sulfur, and Particulate Matter Secondary National Ambient Air Quality Standards (NAAQS) Panel, hereafter referred to as the Panel, met on May 31, 2023, June 28-29, 2023, and September 5-6, 2023, to peer review the EPA's *Policy Assessment for the Review of the Secondary National Ambient Air Quality Standards for Oxides of Nitrogen, Oxides of Sulfur and Particulate Matter (External Review Draft – May 2023)*, hereafter referred to as the PA. The Chartered CASAC approved the Panel's report on September 6, 2023. The CASAC's consensus responses to the charge questions and the individual review comments from the Panel are enclosed. The CASAC recommends that the EPA incorporate the advice provided throughout this report into a Second Draft PA. This Second Draft PA should be brought back to the CASAC for review.

In the PA, the EPA discusses limitations and uncertainties in both the underlying research findings and in the analyses and interpretations done for this PA. While it is important to acknowledge and characterize uncertainty, uncertainty is referred to so often in the PA that it undercuts confidence in the information and conclusions presented. It would be more effective to focus on the most important areas of uncertainty, those that are most relevant to the purpose and recommendations of the PA. It may not be possible to characterize such uncertainties quantitatively, but their relative significance could be characterized. The CASAC suggests that the EPA consider using a more systematic and perhaps quantitative approach to assess uncertainty in the data and analyses used in support of the standard setting process. This approach could help policymakers better understand the degree of uncertainty in the key findings, not just that there is uncertainty, which is always the case.

The CASAC notes that ecosystem effects are largely manifested through atmospheric deposition as evidenced by the material presented in the Integrated Science Assessment (ISA) and this PA. The translation of deposition-based effects to an ambient concentration in air is fraught with difficulties and

complexities. Based on the wording of section 109(b)(2) of the Clean Air Act (CAA), the CASAC sees no reason why NAAQS could not be based on atmospheric deposition. Having a deposition-based standard would be a cleaner, more scientifically defensible approach to standard setting because ecosystem effects are largely characterized and quantified based on atmospheric deposition. The CASAC requests that the EPA reconsider its conclusion that a deposition-based secondary standard cannot be used to protect ecosystems from adverse effects.

A useful and succinct background for the secondary NAAQS for oxides of nitrogen (NO_x), oxides of sulfur (SO_x), and particulate matter (PM) from 1971 to present is given in Chapter 1. It gives an appropriate explanation of the purpose of the PA and its context. It would be useful to add more detail regarding the welfare effects that the current secondary standards are set to protect against. Further explanation is needed of the rationale for the current secondary PM standard, why PM was added to this review of ecological effects, and how the welfare effects evidence considered in the most recent PM review inform the PM standard recommendation in this review. The CASAC recommends that in future reviews, the ecological and welfare effects of a criteria pollutant should be kept together in the same secondary standard review. The EPA should also clarify whether the separate and stand-alone Risk and Exposure Assessment (REA) was not included for only this current secondary NAAQS review process or will not be included for all future NAAQS reviews.

Chapter 2 is well written and documents atmospheric processes, emissions, measurement methods, recent trends, and deposition of nitrogen (N), sulfur (S), and PM. Gaseous ammonia (NH_3) is a critical component of reactive N deposition and its effects on aquatic and terrestrial ecosystems. The PA is inconsistent regarding NH_3 emissions. An upward NH_3 emissions trend with a ~20% increase from 2018-2020 is shown in Figure 2-10. The majority of NH_3 emissions are in the “Other Anthropogenic Sources” category. Emission rates from more specific sub-source categories and their associated uncertainties need to be specified. In the PA, it is noted that fires represent ~5% of total NO_x emissions, but neglects the fact that fires (e.g., wildfires, prescribed and agricultural burning) also emit NH_3 , predominantly during the smoldering phase of combustion. Potential NH_3 emissions during fires need to be estimated. Different monitoring networks are presented separately. It would be helpful to include a map with collocated measurements from all monitoring networks considered. The importance of Clean Air Status and Trends Network (CASTNET) measurements for evaluating NAAQS for ozone, sulfur dioxide (SO_2), nitrogen dioxide (NO_2), and deposition should be emphasized. Information on the current CASTNET network review and potential site elimination conducted by the EPA’s Science Advisory Board should be discussed and referenced. $\text{PM}_{2.5}$ chemical light extinction was proposed as a better indicator than $\text{PM}_{2.5}$ mass as the basis for a visibility-related secondary $\text{PM}_{2.5}$ 24-hour standard during the 2012 PM NAAQS review. Since reconstructed $\text{PM}_{2.5}$ light extinction enhances the influence of NH_4NO_3 and $(\text{NH}_4)_2\text{SO}_4$ relative to most other PM components, it is likely to be a better long-term (annual or multi-year) indicator for N and S deposition than $\text{PM}_{2.5}$ mass.

Chapter 3 describes the current standards and approach. The background information is clearly presented and informative. However, there are a few concepts or topics that are either not addressed or not fully addressed, and should be clarified in a revised PA including: how uncertainty is considered, legacy effects and how they have (or have not) been dealt with in the past, the rationale of adding PM to this review, which secondary effects of PM the review should consider, the rationale for the current secondary standards for PM and what welfare effects they are intended to prevent or limit, and defining the term “quantitative analyses.” The approach described for considering the welfare effects evidence and the risk assessment information for review of the adequacy of the current NO_x , SO_x , and PM secondary standards is relevant and logical.

Chapters 4 and 5 of the PA provide a presentation of the historical body of science supporting the direct and indirect (deposition) secondary effects of NO_x, SO_x, and PM, based on literature (up to ~2017) that was available and used for the 2020 ISA for Oxides of Nitrogen, Oxides of Sulfur and Particulate Matter Ecological Criteria. Chapters 4 and 5 are generally well written and informative. However, despite many useful and informative aspects of the PA, the CASAC has several concerns. Overall, the PA does not adequately represent the state of science to inform a decision by the Administrator on a revised or possibly new NO_x, SO_x, PM secondary standard. The CASAC disagrees with the conclusion that there is generally a lack of quantitative information describing the nutrient enrichment effects of N. The CASAC notes that all the categories considered in the 2020 ISA for atmospheric N deposition effects (terrestrial, wetland, freshwater and estuary) were determined to be causal. The criteria used by the EPA to select a subset of the causal determinations from the ISA for additional risk analysis in PA Chapter 5 are unclear. The PA needs to be revised to clarify these criteria. The PA needs to provide clear reasoning as to why N-driven eutrophication effects from atmospheric N deposition on herbaceous vegetation, lichens and mycorrhizae in terrestrial ecosystems, and eutrophication and toxic blooms in freshwater lakes, are not considered in the quantitative evaluation of risk in Chapter 5. In addition, the PA should be revised to include a quantitative evaluation of the N deposition levels that are protective of estuaries (detailed in the consensus responses).

The literature in the current PA is out of date by over 5 years. Since 2017, there has been substantial new evidence published that would fundamentally change the conclusions of the PA on N deposition effects. The new literature identifies that there are (1) more taxonomic groups affected by elevated N deposition, (2) more national-scale data documenting adverse ecological effects of elevated N and (3) lower levels of N deposition at which causes of ecological adversity are quantified. Taken together, this additional literature supports a greater public welfare concern at lower levels of exposure to N deposition.

Chapter 5 does not present the conclusions by EPA on “risk.” These conclusions need to identify the amount of deposition associated with ecological public welfare “risk” that the EPA believes is acceptable. This information would improve the transparency of the analysis decisions in the PA. Conclusions that identify the levels of deposition that protect U.S. ecosystems from risk, based on the analysis in Chapter 5 could then be clearly linked to options for standards presented in Chapter 7. Currently the links between Chapter 5 and Chapter 7 are not transparent or adequate. Chapter 5 needs to be revised to clearly state which levels of N deposition, S deposition, NO₂, SO₂, and PM are recommended for protection.

The CASAC requests that the PA be revised to convey more information about the role of N deposition in coastal and freshwater ecosystems and to conduct a quantitative risk assessment based on the criteria outlined in the consensus responses. The PA gives too little consideration to the effects on coastal systems, where N causes widespread damage to habitat from eutrophication and harmful algal blooms, and to the effects on freshwater systems, where N can further eutrophication and aggravate toxic cyanobacterial blooms even in low-productivity lakes. Although coastal and freshwater ecosystems are generally impacted by multiple N sources, atmospheric deposition accounts for a significant fraction of “new” N loading to these systems, often ranging from 20 to over 40% of total external loading. Overall, these are highly valued resources with clear impacts associated with elevated N inputs. A range of U.S. estuaries have established N total maximum daily load (TMDL) agreements under the Clean Water Act, and recent quantitative N critical load determinations show widespread exceedances for growth and survival of many tree species on terrestrial landscapes of the coterminous U.S., including National Parks and Class I wilderness areas. These and associated detailed research efforts clearly demonstrate the importance, benefits and need for additional decreases in atmospheric N deposition. In addition to increased attention to ecosystem N deposition and its effects, the CASAC believes there are additional

considerations in Chapters 4 and 5 that will strengthen the PA, including a description and discussion of limitations of indicators of soil impacts of acidification; the effects of N+S deposition on freshwater acidification; a clear statement of limitations and assumptions associated with the applications of steady-state critical loads models of acidification; and climate and legacy impacts of atmospheric deposition. Finally, the PA needs to make it clear why effects described in various sections of Chapter 4 are important. Effects do not have to be fully quantified to be confident that they are causing associated public welfare effects.

In Chapter 6, an extensive evaluation of the relationships between air quality metrics and deposition of N and S to ecosystems is presented. In the PA, there are numerous scatterplot matrices showing relationships between measured PM_{2.5} constituents from various monitoring networks and measured wet N and S deposition, and between measured PM_{2.5} constituents and modeled values of N and S deposition. The CASAC recommends that the EPA harmonize their presentation, and provide more discussion of the most useful relationships.

In the national scale sites-of-influence analysis, EPA uses the National Oceanic and Atmospheric Administration (NOAA) Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model to establish quantitative relationships between ambient pollutant concentrations at upwind source sites (or “sites of influence”) and deposition of N and S at downwind sites. The CASAC recognizes the fundamental difficulties of finding quantitative relationships between air quality concentrations and deposition, and acknowledges that the problem of using atmospheric measurements to predict deposition has not been adequately researched or documented in the peer-reviewed literature. This shortcoming required the EPA to develop the Ecoregion Air Quality Metric (EAQM) methodology in-house.

All CASAC members agree that the description of the EAQM approach and methodology lacks sufficient detail; that many of the parameters and assumptions need to be explained and justified; and that the EPA should perform sensitivity analyses to help quantify the uncertainty associated with this approach. The CASAC recommends that the EPA expand and document the EAQM model description, provide output in a format that facilitates interpretation of results, and add additional explanation of data that do not conform to expected relationships. The CASAC provides detailed comments on the EAQM method for EPA’s consideration in its consensus responses.

The CASAC does not have consensus on its response to the technical approach in Chapter 6. The EAQM results presented in Chapter 6 are used in Chapter 7 to support the secondary NAAQS recommendations. Since the EAQM modeling work has not been peer reviewed, the results should be viewed with skepticism. However, some CASAC members find that the EAQM results are still useful since there are limited analyses available that compare SO₂, NO₂, and PM_{2.5} design values to S and N deposition values. Other CASAC members find that the EAQM results are not scientifically sound and should not be used to inform the secondary standard recommendations for SO₂, NO₂, and PM_{2.5}. Assuming the EPA continues to use the EAQM approach, the CASAC has several comments and recommendations detailed in the consensus response. If this approach is continued, updates to the EAQM analysis based on the CASAC’s advice should be included in the Second Draft PA.

On July 24, the EPA released a memo titled “Correction to Errors in Six Figures of Chapter 6 of the Policy Assessment for the Review of the Secondary National Ambient Air Quality Standards for Oxides of Nitrogen, Oxides of Sulfur and Particulate Matter, External Review Draft.” The EPA followed this up on July 27 with a memo that also provided a table of correlation coefficients for correlations of deposition estimates with various air quality metrics as well as attachments with the corrected figures embedded in the corrected text of the PA. The corrected figures address some concerns raised by the

CASAC, but do not address other concerns, such as the highly pronounced bifurcation in the plotted relationships. The Eastern versus Western ecoregion groupings is not mentioned in the PA, including in the corrected text. Without incorporation of this analysis into the PA itself, the utility of this distinction between ecoregions is unclear.

The CASAC does not find the information on potential welfare effects of N and S in ambient air and in deposition reviewed in the ISA and PA to be adequately summarized (as noted above and detailed in the consensus responses). The CASAC urges the EPA to adopt secondary standards based on deposition rather than exposure given the strong, decades-long evidence for deposition-related effects on ecosystems. To protect public welfare against the effects of NO_x, SO_x, and PM, the CASAC believes that the secondary standard should be based on the strongest existing evidence for ecological effects of these pollutants. Several decades of research show that ecosystem effects are largely manifested through atmospheric deposition as presented in the 2020 ISA. The ISA reviews N and S deposition effects on six categories of ecosystem structure and function in terrestrial, wetland, freshwater, and estuarine ecosystems and all six categories were determined to be causal in the ISA. In contrast, relating atmospheric deposition-based effects to ambient concentrations is fraught with difficulties. The CASAC thus recommends that direct atmospheric deposition standards be considered in future reviews. Direct atmospheric deposition of S and N pollutants is a clear and straightforward indicator of “the presence of these pollutants in the ambient air.”

Nonetheless, the CASAC provides the following recommendations for alternative standards based on air concentrations that should be protective of direct and deposition-related effects. The EPA should incorporate these recommended alternative standards into the Second Draft PA. The CASAC provides detailed justification for each recommendation in its consensus comments.

The CASAC has consensus regarding the 3-hour SO₂ standard for direct effects. The current SO₂ secondary standard of 0.5 ppm (3-hour maximum not to be exceeded more than once per year) is adequate to protect against the direct effects of oxides of S on plants and lichens and should be retained without revision.

The CASAC also has consensus regarding the annual NO₂ standard for direct effects. The current annual secondary NO₂ standard of 53 ppb is adequate to protect against the direct welfare effects of N oxides in ambient air on plants and lichens and should be retained without revision.

All of the CASAC members,* except one, recommend the following standards to address deposition-related effects of N and S:

- Annual PM_{2.5} standard - The current secondary PM_{2.5} standard is inadequate to protect ecosystems against the deposition-related effects of N and S, and a secondary annual PM_{2.5} standard in the range of 6-10 µg/m³ is recommended. A new total N PM_{2.5} indicator is recommended to achieve a better measure of total reactive N deposition, including ammonium deposition, during the next review.
- Annual NO₂ standard - The current annual secondary NO₂ standard of 53 ppb does not provide sufficient protection against the deposition-related welfare effects of N and is therefore inadequate. The secondary annual NO₂ standard should be revised to <10-20 ppb.

* This also includes all of the CASAC Panel members, except for the one CASAC member and one additional panel member.

- Annual SO₂ standard – The current SO₂ standard does not afford adequate protection against the deposition-related effects of S on ecosystems. A new secondary annual SO₂ standard is recommended in the range of 10-15 ppb to preclude the possibility of returning to deleterious deposition values as seen associated with the emergence of high annual average SO₂ concentrations near industrial sources in 2019, 2020, and 2021 (Figure 2-25).
- 24-hour PM_{2.5} standard – The current 24-hour standard is not adequate to protect against short-term episodic events. A 24-hour PM_{2.5} secondary standard of 25 µg/m³ or a level of 20-25 deciviews based on a PM_{2.5} chemical light extinction indicator is recommended.

However, one CASAC member finds that the historical Total Deposition (TDep) values demonstrate that the current primary NAAQS for SO₂, NO₂, and annual PM_{2.5} provide adequate protection against “long-term” annual S and N deposition-related effects; therefore, the secondary NAAQS for SO₂, NO₂, and annual PM_{2.5} should be set equal to the current primary NAAQS for SO₂, NO₂, and annual PM_{2.5}. Please see Appendix A for details on the dissenting viewpoint.

The CASAC wishes to highlight that virtually all the research in this PA depends heavily on data from our various national monitoring networks: National Air Monitoring Stations (NAMS)/State and Local Air Monitoring Stations (SLAMS), Chemical Speciation Network (CSN), Interagency Monitoring of Protected Visual Environments (IMPROVE), CASTNET, National Atmospheric Deposition Program (NADP), and more. Without these data, there would be no scientific basis for establishing new standards. Indeed, these networks were established in part to provide data for such research, as well as for assessing health and welfare impacts from air pollution, providing data for model evaluation, tracking pollutant trends, evaluating emerging pollutants, and assessing compliance with ambient standards and emission controls. Determining effects of climate change has become another critical challenge requiring long-term measurements from robust and distributed networks. The importance of these networks in support of the EPA’s mission of protecting and managing air quality is indisputable. Despite their value, these networks are faced perennially with uncertain funding and cutbacks that threaten the continuity of the critical data stream they provide. In May 2022, the EPA announced the termination or suspension of a number of sites associated with major national air quality monitoring programs, including CASTNET, National Trends Network (NTN), Ammonia Monitoring Network (AMoN) and Mercury Deposition Network (MDN) sites. Now in 2023, the EPA is proposing to close additional sites from CASTNET, which would leave major gaps in the understanding of air pollution under current and future air and climate policies. Rural monitoring sites in these networks provide air quality data in locations where state and local monitors rarely exist, making them an important resource for rural, low-income communities that often lack the funding and expertise to operate their own monitoring programs. Robust long-term air quality networks are critical to our understanding and protection of people, underserved communities, and ecosystems during a period of changing climate. It is essential that they are supported and maintained to continue to support the protection of public health, natural resources, and sustainable economic growth. The CASAC recommends that EPA prioritize network funding, for these and the other networks that come under its jurisdiction.

The CASAC appreciates the opportunity to provide advice on the PA and looks forward to the agency's response.

Sincerely,

/s/

Dr. Elizabeth A. (Lianne) Sheppard, Chair
Clean Air Scientific Advisory Committee

Enclosures

NOTICE

The Clean Air Scientific Advisory Committee (CASAC) is a chartered federal advisory committee, operating under the Federal Advisory Committee Act (FACA; 5 U.S.C. § 10). The committee provides advice to the Administrator of the U.S. Environmental Protection Agency on the scientific and technical bases of the National Ambient Air Quality Standards. The findings and recommendations of the committee do not represent the views of the Agency, and this document does not represent information approved or disseminated by EPA. The CASAC reports are posted on the EPA website at:

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U.S. Environmental Protection Agency Clean Air Scientific Advisory Committee

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**U.S. Environmental Protection Agency
Clean Air Scientific Advisory Committee
Oxides of Nitrogen, Oxides of Sulfur, and Particulate Matter (PM) Secondary
National Ambient Air Quality Standards (NAAQS) Panel**

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**Consensus Responses to Charge Questions on the EPA's
*Policy Assessment for the Review of the Secondary National Ambient Air Quality Standards for
Oxides of Nitrogen, Oxides of Sulfur and Particulate Matter (External Review Draft – May 2023)***

Overarching Comments on Uncertainty

Starting in Chapter 1, limitations and uncertainties are discussed in the Policy Assessment (PA) both in the underlying research findings and in the analyses conducted and interpretations presented. While it is important to acknowledge and characterize uncertainty, the document refers to uncertainty so often that this repetitiveness undercuts confidence in the information and conclusions presented. It would be more effective to focus on the most important areas of uncertainty that are most relevant to the purpose and recommendations of the PA. It may not always be possible to characterize such uncertainties quantitatively, but their relative significance could be characterized.

When considering the policy question of what standards would be requisite to protect ecosystems from harmful welfare effects, it is important to recognize that uncertainty can work two ways. There is risk of over controlling, i.e., setting a standard that is too stringent. There is also risk of under controlling, i.e., setting a standard that is too lenient. An overemphasis on uncertainty is likely to cause an error toward under controlling. The manner in which uncertainty is emphasized in the PA tends to undercut recognition of already well-established scientific understanding of the effects under consideration.

Over time, harmful effects at lower and lower levels of atmospheric deposition on ecosystems continue to be identified. Comparing the last two Integrated Science Assessments (ISAs), the number of causal relationships supported by the literature has increased. This trend of finding additional effects as research progresses suggests that accounting for uncertainty should favor the more protective end of the range supported by the current evidence.

One major area of uncertainty is the translation of ambient concentrations to atmospheric deposition levels presented in Chapter 6. Specific suggestions regarding those analyses are included in the Chapter 6 consensus response. Previous Risk and Exposure Assessments (REAs) and PAs have included new analyses to apply results available in the literature to the specific policy questions raised by the standard setting process. It is not clear whether the analyses in Chapter 6, specifically the those for the Ecoregion Air Quality Metric (EAQM) methodology, are adequate for supporting standard setting. The CASAC does not have consensus on this point. Some members think that the modeling approach used in Chapter 6 is too simplified and the results are too weak to provide a defensible basis for standard setting. Other members believe that the results of the EPA's analyses can be used in a limited way when combined with other evidence presented in the PA to identify a range of concentrations associated with atmospheric deposition levels below identified thresholds.

The CASAC suggests that the EPA consider using a more systematic and perhaps quantitative approach to assessing uncertainty in the data and analyses used to support the standard setting process. This could help policymakers better understand the degree of uncertainty in the key findings, not just that there is uncertainty, which is always the case. An example to consider is how uncertainty is addressed both qualitatively and quantitatively by the Intergovernmental Panel on Climate Change in their Climate Change 2021 report. Another recent example of quantitative uncertainty estimates (and qualitative discussion) for sulfur (S) and nitrogen (N) critical loads (CLs) for growth and survival of U.S. tree species is Pavlovic et al. (2023), as referenced in individual panel member comments from Dr. Charles Driscoll, Jr.

Overarching Comments on Atmospheric Deposition-Based Standards

For the history of the National Ambient Air Quality Standards (NAAQS), standards of criteria air pollutants have been based on ambient air concentrations. Nevertheless, ecosystem effects are largely manifested through atmospheric deposition as evidenced by the material presented in the ISA and this PA. The EPA goes through an extensive evaluation in Chapter 6 of the PA of translations of deposition-based effects to concentrations of nitrogen dioxide (NO₂), sulfur dioxide (SO₂) and particulate matter (PM) in ambient air. Under section 109(b)(2) of the Clean Air Act (CAA), a secondary standard must “specify a level of air quality the attainment and maintenance of which, in the judgment of the Administrator, based on such criteria, is requisite to protect the public welfare from any known or anticipated adverse effects associated with the presence of [the] pollutant in the ambient air.” Based on this wording of the CAA, the CASAC sees no reason why NAAQS could not be based on atmospheric deposition. The wording “level of air quality” is sufficiently broad that it could and should include deposition. Ambient is defined as relating to the immediate surroundings. Atmospheric deposition should be understood as a component of “ambient air.” It is a measure of pollution from the ambient air that has landed on the ground or other surfaces. Having an atmospheric deposition-based standard would be a cleaner, more defensible approach because ecosystem effects are largely characterized and quantified based on deposition. Moreover, the translation of deposition-based effects to an ambient concentration is fraught with difficulties and complexities as articulated in the consensus responses for Chapter 6, including effects of elevation, land cover, meteorological conditions, proximity to emission sources among other factors. The CASAC requests that the EPA reconsider its conclusion that an atmospheric deposition-based secondary standard cannot be used to protect ecosystems from adverse effects.

Chapter 1 – Introduction

To what extent does the Panel find that the information in Chapter 1 is clearly presented and that it provides useful context for the review?

Chapter 1 gives a useful and succinct background for the secondary NAAQS for oxides of nitrogen (NO_x), oxides of sulfur (SO_x), and particulate matter (PM) from 1971 to present. It gives an appropriate explanation of the purpose of this PA and its context.

It would be useful to add more detail regarding the welfare effects that the current secondary standards are set to protect against. Further explanation is needed of the rationale for the current secondary PM standard. The PA does not make it clear to what extent ecological effects were considered in the last PM review. The CASAC requests a more thorough explanation of why PM was added to this review of ecological effects. There should also be direct discussion of how the welfare effects evidence considered in the most recent PM review inform the PM standard recommendation in this review. The CASAC recommends that in future reviews, the ecological and welfare effects of a criteria pollutant should be kept together in the same secondary standard review.

As further context for this review, the Acid Rain Program under the 1990 Clean Air Act Amendments (CAAA) should be described. This program was motivated by recognition of the harm caused by acid deposition; that recognition has helped reduce precursor emissions, especially SO₂. That Congress included acid rain in the Amendments is strong support that adverse public welfare effects of acid deposition were recognized at that time.

As noted above, the CASAC recommends that the EPA revisit their interpretation that the CAA allows only for a standard based on an ambient air concentration measurements. The CASAC finds that a direct atmospheric deposition standard is the most scientifically defensible approach for protecting ecosystems. For purposes of a scientific evaluation of pollutant effects on public welfare, ambient air quality also includes any portion of the atmosphere in which pollutants are present that may reasonably be anticipated to cause or contribute to effects on public welfare. It should be noted that a feasibility assessment of setting a deposition standard was mandated under the 1990 CAA Amendments and was conducted in the 1990s. Additionally, in 2012, a previous CASAC panel supported a proposed secondary standard that used an aquatic acidification index (AAI) based on acid neutralizing capacity (ANC) for protection of aquatic ecosystems that incorporated estimates of acidic deposition of S and N compounds and that also considered the role of reduced nitrogen compounds (NH_x). It should be made clear whether secondary standards have to be the same throughout the country or whether they can vary for locations with different sensitivities to the effects of the pollutants.

The term “quantitative analyses” is referred to in Chapter 1. It appears that this pertains to analyses that previously were conducted in the REA. It would be helpful to be explicit about this “quantitative analysis” as interested parties may wonder why the REA was not conducted for this assessment. Also, it is not clear if the separate and stand-alone REA was not conducted for only this current secondary NAAQS review process or if this change will occur for all future EPA NAAQS reviews.

In Chapter 1, it is noted that this PA “is written to be understandable to a broad audience.” This is a laudable goal, which is difficult to achieve. This PA would benefit from revision by an external editor with technical, science, and policy expertise to make it more readable to the broader scientific community and the public.

Chapter 2 – Air Quality

To what extent does the Panel find that the information in Chapter 2 is clearly presented and that it provides useful context for the review?

Chapter 2 is well written and documents atmospheric processes, emissions, measurement methods, recent trends, and deposition of N, S, and PM. The following comments are suggestions for improvement.

2.1 Atmospheric Transformation of N, S, and PM species

In this section, atmospheric processes are briefly described without acknowledging the non-linear relationships between the precursor gas concentrations and the rate or extent of secondary aerosol formation. It would be informative to address the complex chemistry of nitrate (NO_3^-) and sulfate (SO_4^{2-}), the formation pathways, and changes in equilibrium with gaseous ammonia (NH_3), nitric acid (HNO_3), and sulfuric acid (H_2SO_4) under different temperature and relative humidity conditions.

An overview of the acid deposition process should be provided. Oxidation of SO_2 to H_2SO_4 occurs largely in the aqueous phase of clouds, mainly through reaction with hydrogen peroxide (H_2O_2); this oxidation pathway thus couples the gas-phase photochemical system for SO_2 oxidation. In contrast, oxidation of nitric oxide (NO) and NO_2 to HNO_3 occurs in the gas phase as well as via heterogenous

hydrolysis of dinitrogen pentoxide (N_2O_5); gaseous HNO_3 is rapidly and nearly completely taken up by cloud water. Deposition of NO_3^- and SO_4^{2-} occurs predominantly through precipitation.

Sulfur oxides (SO_x) are present in multiple forms: SO_2 (g), bisulfite ion (HSO_3^-) (aq), sulfite ion (SO_3^{2-}) (aq), H_2SO_4 (aq), hydrogen bisulfate ion (HSO_4^-), and sulfate ion (SO_4^{2-}). The EPA should clarify in the PA whether or not primary SO_4^{2-} emitted from combustion sources is part of the SO_x emissions. Atmospheric SO_4^{2-} present in the forms of ammonium sulfate ($(\text{NH}_4)_2\text{SO}_4$) and ammonium bisulfate (NH_4HSO_4) depend on the available NH_3 under different environments.

2.2 Sources and Emissions of N, S, and PM Species

Gaseous NH_3 is a critical component of reactive N deposition and its effects on aquatic and terrestrial ecosystems. The PA is inconsistent regarding NH_3 emissions. An upward NH_3 emissions trend is shown in Figure 2-10, with a ~20% increase from 2018-2020. The majority of NH_3 emissions are in the “Other Anthropogenic Sources” category. Emission rates from more specific sub-source categories and their associated uncertainties need to be specified.

Section 2.2.3 (NH_3 Emissions) notes that ~2% of total NH_3 emissions attributed to mobile sources in the National Emissions Inventory (NEI) may be underestimated. On-road and off-road vehicle emissions warrant further discussion. Relevant studies should be cited regarding catalytic converters that reduce NO_x but may produce NH_3 emissions. The use of urea to treat diesel exhaust from trucks may also lead to an increase in NH_3 emissions.

It is noted in the PA that fires represent ~5% of total NO_x emissions, but the fact that fires (e.g., wildfires, prescribed and agricultural burning) also emit NH_3 is neglected, predominantly during the smoldering-phase of combustion. Potential emissions during fires need to be estimated.

2.3 Monitoring Ambient Air Concentrations and Deposition of N, S, and PM

Different monitoring networks, i.e., NO_x , SO_x , $\text{PM}_{2.5}$, Clean Air Status and Trends Network (CASTNET), National Atmospheric Deposition Program (NADP)/National Trends Network (NTN), and Ammonia Monitoring Network (AMoN) are presented separately. It would be helpful to include a map with collocated measurements like those shown in Figure 6-2 for the 27 collocated Interagency Monitoring of Protected Visual Environments (IMPROVE), CASTNET, and NADP/NTN sites. Inter-network differences in sampling and analysis should be noted and discussed. The monitoring network for NH_3 is sparsely and unevenly distributed with most monitors located in the eastern U.S. Note that areas with the highest NH_3 emissions (e.g., Iowa and Southern Minnesota) do not have NH_3 monitors. Satellite observations could be applied to fill in the gaps. The lack of NH_3 measurements in the Midwest and Plains states increases uncertainties for N deposition and critical load estimates.

The importance of CASTNET measurements for evaluating the NAAQS for O_3 , SO_2 , NO_2 , and deposition should be emphasized. Information on the current CASTNET network review and potential site elimination conducted by the EPA’s Science Advisory Board should be discussed and referenced.

2.4 Recent Ambient Air Concentrations and Trends

Additional graphs and discussion could be added to illustrate temporal variations of secondary SO₂ standard design values (second highest daily 3-hour average) and to display data distributions between maximum three hourly average concentration and annual average SO₂ at selected sites.

The PM_{2.5} chemical composition shown in Figure 2-26 should include measured or calculated NH₄⁺; change organic carbon (OC) to organic mass (OM) with an appropriate multiplier to account for unmeasured oxygen and hydrogen; and include the “unidentified material” (possibly liquid water, artifact, etc.) to infer potential uncertainties. It would be informative if the diameter of each pie chart in Figure 2-26 was proportional to PM_{2.5} mass to illustrate spatial concentration variability.

PM_{2.5} chemical light extinction was proposed as a better indicator than PM_{2.5} mass as the basis for a visibility-related secondary PM_{2.5} 24-hour standard during the 2012 PM NAAQS review. Since reconstructed PM_{2.5} light extinction enhances the influence of NH₄NO₃ and (NH₄)₂SO₄ relative to most other PM components, it is likely to be a better long-term (annual or multi-year) indicator for N and S deposition than total PM_{2.5} mass. Recent (2018) EPA guidance on tracking progress under the “Regional Haze Rule” screens out episodic influences from wildfires and dust storms to focus on the 20% most (anthropogenically) impaired days, placing even greater emphasis on N and S compounds. In future reviews it would be instructive to include spatial and temporal trends of PM_{2.5} light extinction composition, which could be calculated for the ~300 sites in the urban Chemical Speciation Network (CSN) and rural IMPROVE networks. Also, future reviews could examine how the most impaired days would influence the PM_{2.5} light extinction indicator for both ecological and visibility effects of N and S compounds.

Figures for spatial distributions and long-term trends of NH₄⁺ should be presented and compared with trends for NO₃⁻ and SO₄⁼. In future reviews, EPA is encouraged to consider routinely measured (or calculated) PM_{2.5} components of NH₄⁺, NO₃⁻, and SO₄⁼ (or total N and total S) in the U.S long-term networks as indicators to represent N and S deposition.

2.5 N and S Deposition

Flow diagrams for estimating dry deposition differ between Figures 2-36 and 2-37; these two figures could be combined to illustrate data sources for calculating total deposition. Uncertainties associated with model estimated dry deposition should be clarified. To the extent that modeled Community Multiscale Air Quality (CMAQ) concentrations are used to estimate dry deposition, uncertainties of dry deposition flux might be obtained by comparing CMAQ concentrations with measurements.

Total N and S deposition rates from 2019-2021 are shown in Figures 2-38 and 2-39. It would be helpful to provide figures that separate wet and dry deposition to distinguish their relative magnitudes. Similar graphs should also be provided indicating percent distributions for reduced N. Causes of the increase of total NH_x (NH₃ + NH₄⁺) deposition by wet and dry deposition over the past 20 years should be clarified.

Table 2-26 is a good example that demonstrates changes in total oxidized and reduced N deposition by region between the 2000-2002 and 2019-2021 periods. More discussion is warranted to highlight the non-linear nature of nitrate chemistry.

Chapter 3 – Current Standards and Approach

To what extent does the Panel find that the background information in sections 3.31 and 3.2 to be clearly presented and provide useful context for the review?

The background information in Chapter 3 is clearly presented and informative. However, there are a few concepts or topics that are either not addressed, or not fully addressed, and should be clarified. A few of these are discussed below and some may not need more than a phrase or sentence to be added, especially if they are more fully addressed in other places in the document. The term “uncertainty” appears frequently throughout the document without a clear description or definition. In this background section, it would be helpful to describe how uncertainty has been considered and placed in the context of levels of certainty that have been identified. For example, a paragraph that details some of the largest model and data uncertainties that led to not adopting a new secondary standard to protect against deposition-related welfare effects after earlier reviews is relevant. There is an absence of a discussion on legacy effects and how they have (or have not) been addressed in the past. For example, it is not clear how the secondary standards for future “new” deposition of sulfates, nitrates, and PM (and, perhaps, ammonia and ammonium) would be addressed in light of the historic baseline of large cumulative deposition of these pollutants to aquatic and terrestrial ecosystems. Background information should be added on why PM has been added to this review, and which parts of the secondary effects of PM the review should consider, should not consider, and why. It would be helpful to add discussion of the rationale for the current secondary standards for PM and what welfare effects they are intended to prevent or limit. The term “quantitative analyses” should be defined or clarified.

What are the Panel’s views on the general approach described in section 3.3 for considering the welfare effects evidence and the risk assessment information for review of the adequacy of the current NO₂, SO₂ and PM secondary standards?

The approach described is relevant and logical. Important advances in this review include the focus on the deposition-related effects, and the addition of PM.

Chapter 4 – Nature of Welfare Effects and Chapter 5 – Exposure Conditions Associated with Effects

To what extent does the Panel find the summary of the current evidence on the ecosystem effects of oxides of nitrogen, oxides of sulfur and particulate matter in ambient air, including consideration of both the evidence for direct effects of the pollutants in ambient air and for the effects of the associated atmospheric deposition into aquatic and terrestrial ecosystems, to be technically sound and clearly communicated? What are the Panel’s views on the consideration of the public welfare implications, in light of judgments and conclusions made in NAAQS reviews regarding effects on the public welfare?

Does the Panel find the presentation of information and quantitative analyses related to welfare effects of oxides of nitrogen, oxides of sulfur and particulate matter to be technically sound and clearly communicated? What are the Panel’s views on the quantitative analysis of aquatic acidification risks, including the choice of analysis approach and spatial resolutions? Does the document appropriately characterize the results of the analysis, including key limitations and associated uncertainties?

Chapters 4 and 5 of the PA provide a presentation of the historical body of science supporting the direct and indirect (deposition) secondary effects of NO_x, SO_x, and PM, based on literature that was available and used for the 2020 ISA (~2017). Chapters 4 and 5 are generally well written and informative. The CASAC has chosen to combine comments on these two chapters, given that Chapter 4 presents the nature of welfare effects, while Chapter 5 builds on that foundation to define the exposure conditions of those same effects in a risk assessment. Many of the comments provided apply to both chapters unless otherwise noted.

Despite many useful and informative aspects of the PA, the CASAC has several concerns. Overall, the PA does not adequately represent the state of science to inform a decision by the Administrator on a revised or possibly a new NO_x, SO_x, PM secondary standard based on the literature through 2017. First and foremost, the CASAC disagrees with the conclusion in Chapter 5 that there is generally a lack of quantitative information describing the nutrient enrichment effects of N. The CASAC notes that all the categories considered in the 2020 ISA for atmospheric N deposition effects (terrestrial, wetland, freshwater and estuary) were determined to be causal (Figure ES-3, 2020 ISA). The criteria used by the EPA to select a subset of the causal determinations from the ISA for additional risk analysis in Chapter 5 of the PA are unclear. The PA needs to be revised to clarify these criteria. Given that the ISA is intended to guide the PA, the CASAC finds this inconsistency disconcerting, particularly considering the poorly described reasoning for these decisions in the PA. The quantitative analysis in the PA focuses on freshwater acidification, terrestrial acidification, and N deposition effects on tree growth. The PA needs to also provide clear reasoning as to why N-driven eutrophication effects from atmospheric N deposition on herbaceous vegetation, lichens and mycorrhizae in terrestrial ecosystems, and eutrophication and toxic blooms in freshwater lakes, are not considered in the quantitative evaluation of risk in Chapter 5. In addition, the PA should be revised to include a quantitative evaluation of the N deposition levels that are protective of estuaries (outlined below).

Second, the literature in the current PA is out of date by over 5 years (the literature cut-off date is May 2017). Since May 2017, there has been substantial new evidence published that will fundamentally change the conclusions of the PA on N deposition effects. The new literature identifies that there are (1) more taxonomic groups affected by elevated N deposition, (2) more national-scale data documenting adverse ecological effects of elevated N and (3) lower levels of N deposition at which causes of ecological adversity are quantified. Taken together, this additional literature supports a greater public welfare concern at lower levels of exposure to N deposition.

Third, Chapter 5 does not present the conclusions by the EPA on “risk.” These conclusions need to identify the amount of deposition associated with ecological public welfare “risk” that the EPA believes is acceptable. This information would improve the transparency of the analysis decisions in the PA. Conclusions that identify the levels of deposition that protect U.S. ecosystems from risk, based on the analysis in Chapter 5 could then be clearly linked to options for standards presented in Chapter 7. Currently the links between Chapter 5 and Chapter 7 are not transparent or adequate. Chapter 5 needs to be revised to clearly state which levels of N deposition, S deposition, NO₂ and SO₂ are recommended for protection.

The CASAC requests that the PA be revised to convey more information about the role of N deposition in coastal and freshwater ecosystems and to conduct a quantitative risk assessment based on the criteria outlined in these consensus responses. In the PA, the EPA gives too little consideration to the effects on coastal systems, where N causes widespread damage from eutrophication and harmful algal blooms, and to the effects on freshwater systems, where N can further eutrophication and aggravate toxic

cyanobacterial blooms even in low-productivity lakes (Howarth, 1988; NRC, 2000; Rabalais et al., 2002; Paerl and Piehler, 2008; Howarth, 2022). Admittedly, coastal and freshwater ecosystems are generally impacted by multiple N sources, including wastewater effluent, urban and agriculture runoff, as well as atmospheric deposition which complicates assessments (Driscoll et al., 2003). However, atmospheric deposition accounts for a significant fraction of “new” N loading to these systems, often ranging from 20 to over 40% of total external loading (Paerl et al., 2002). Overall, these ecosystems are highly valued resources with clear impacts associated with elevated N inputs (Paerl, 2002; Fenn et al., 2003). As outlined below, a range of U.S. estuaries have established N total maximum daily load (TMDL) agreements under the Clean Water Act.

In addition, recent quantitative N critical load determinations show widespread exceedances for growth and survival of many tree species on terrestrial landscapes of the coterminous U.S., including National Parks and Class I wilderness areas (Pavlovic et al., 2023; M. Bell NPS comments to CASAC). These and other research efforts clearly demonstrate the importance, benefits and need for additional decreases in atmospheric N deposition to protect ecosystems broadly across the U.S.

Beyond increased attention to ecosystem N deposition and its effects, the CASAC believes there are additional considerations in Chapters 4 and 5 that will strengthen the PA, including a description and discussion of limitations of indicators of soil impacts of acidification; a clear statement of limitations and assumptions associated with the applications of steady-state critical loads models of acidification; and climate and legacy impacts of atmospheric deposition.

1. N effects

There are benefits and disbenefits of enhanced atmospheric N deposition associated with human activities. Benefits include fertilization of crops and trees and the potential for improved sequestration of carbon in soils and plant biomass. Disbenefits include reductions in microbial and plant species diversity; shifts in mycorrhizal associations; alterations in ecosystem function, including changes in soil carbon pools, enhanced mineralization of organic N, nitrification, nitrate leaching and nitrous oxide production; eutrophication of N-limited freshwaters, including increased toxin production and release from harmful algal blooms (HABs); eutrophication of coastal waters involving loss of submerged aquatic vegetation, and development of hypoxic conditions that severely degrade coastal habitats. In general, there is variability across landscapes where benefits and disbenefits of atmospheric N deposition are realized. Atmospheric N deposition is generally considered a net benefit to croplands, although Lobell and Burney (2021) observed damaging effects of ambient N dioxide to maize and soybeans in the Midwest. In contrast, disbenefits likely outweigh benefits associated with atmospheric N in extensively managed lands (forests, grasslands), as well as freshwaters and coastal waters. Urban ecosystems experience particularly elevated atmospheric N deposition (Decina et al., 2017; Conrad-Rooney et al., 2023). While urban forests in the northeastern U.S. have increased soil N availability and rates of N uptake by trees compared to rural forests (Caron et al., 2023), it is not clear if urban terrestrial ecosystems are adversely impacted by elevated N deposition, but this is a topic of increased research and management interest.

Terrestrial ecosystems have evolved over geologic time under low atmospheric N deposition and the disbenefits of N inputs in extensively managed lands likely stem from this condition. These potential disbenefits of N inputs in ecosystems are broad-based and comprehensive. They are complex and challenging, but also clearly affect the fundamental structure and function of terrestrial, freshwater, and marine ecosystems with strong linkages to a host of important ecosystem services. The sheer extent and

magnitude of ecosystem effects of elevated human-generated N inputs is indicative of the breadth and importance of this environmental problem. EPA needs to adequately consider these effects and take a leadership role in mitigating this agent of disturbance. The CASAC recommends that the EPA strengthen the justification in the PA for the need to limit N deposition across diverse landscapes to mitigate ecosystem effects of elevated N inputs. Additional detail to support this justification is provided below.

An important consideration of atmospheric N deposition is legacy effects. Terrestrial ecosystems accumulate N, and this N resides in different pools with slow or fast turnover rates. Gilliam et al. (2019; 2023) developed a conceptual framework of the hysteresis in terrestrial ecosystem response to first increases and then decreases in atmospheric N deposition. Wood and soil organic matter have slow N turnover rates, while microbial biomass and mineral soil exhibit rapid N turnover rates. Ecosystem responses that are governed by N pools with slow turnover rates will show greater hysteresis. In contrast, ecosystem responses governed by N pools with rapid turnover rates will show less hysteresis. For example, nitrate leaching is governed by fast turnover N pools and will generally respond rapidly to decreasing N deposition. Due to these legacy effects some of the impacts of atmospheric N deposition appear to be realized rapidly while others manifest over multiple decades. Likewise following management actions some effects are mitigated relatively rapidly while others linger for extended periods. It would be useful for the EPA to consider incorporating this conceptual model into the PA to integrate the terrestrial and aquatic effects of atmospheric N deposition and address the varying timescales for these effects to be realized or mitigated.

1.1 Recent terrestrial effects (trees, understory, soils, microbial and mycorrhizal)

As mentioned, numerous studies have demonstrated adverse effects of atmospheric N deposition to extensively managed terrestrial ecosystems. This body of research encompasses studies relying on a variety of approaches to come to these conclusions, including experimental addition and removal studies (Patel et al., 2020; Gilliam et al., 2020), time-series studies (Driscoll et al. 2016), gradient analysis (Aber et al., 2003; Baron et al., 2011), paleoecological studies (Baron et al., 2011), and modeling (Shao et al., 2020, 2021). Elevated atmospheric N deposition has been shown to influence a host of terrestrial ecosystem characteristics, including soils (Gilliam et al., 2019), mycorrhizal associations (Averill et al., 2018, Jo et al., 2019), understory vegetation (Simkin et al., 2016; Clark et al., 2019; Gilliam et al., 2016; McDonnell et al., 2020), overstory tree growth and survival (Thomas et al., 2009; Horn et al., 2018), lichens (Geiser et al., 2019), and surface water leaching (Aber et al. 2003; Baron et al. 2011). Comprehensive critical loads for N deposition have been evaluated for trees (Pavlovic et al., 2023) and understory vegetation (McDonnell et al., 2020). Moreover, critical loads have been determined showing widespread exceedances for National Parks and Class I wilderness areas (see M. Bell NPS comments to CASAC). The CASAC refers the EPA to the analysis of Pavlovic et al. (2023) in which machine learning was used to analyze the effects of atmospheric N and S deposition on the growth and survival of various tree species using data from the United States Forest Service (USFS) Forest Inventory and Analysis (FIA). This machine learning approach provides a more robust analysis quantifying species-specific responses of tree growth and survival to recent levels of atmospheric S and N deposition than had been previously determined for the coterminous U.S. Moreover, these investigators characterized uncertainty limits for tree species' critical loads of N and S. Machine learning can also be used to characterize the mediating factors that influence the response of tree species to atmospheric deposition. This and other recent analyses indicate ongoing effects from atmospheric N and S that have been quantified and well-characterized for many forest ecosystems (e.g., Clark et al., 2017, 2019; Gilliam et al., 2016, 2018, 2020; Walter et al., 2017) across the U.S.

1.2 Freshwaters

The role of N in eutrophication in lakes is more nuanced than for coastal waters, as discussed below. Many leading freshwater ecologists have long maintained that phosphorus is the key driver of freshwater eutrophication (Schindler et al., 2008). In comparison to coastal marine ecosystems where N limitation is widely prevalent, phosphorus limitation of primary production is dominant in many freshwater lakes (Howarth & Marino 2006; Elser et al., 2007; Schindler et al., 2008). However, a growing body of evidence shows that for many eutrophic lakes (Fenn et al., 2003), N may be limiting or co-limiting with phosphorus, in part due to accumulation of legacy phosphorus in watersheds and sediments. As a result, scientists have increasingly called for controlling inputs of both phosphorus and N to lakes (Howarth and Paerl, 2008; Conley et al., 2009; Paerl, 2009; Lewis et al., 2011; Paerl et al., 2016; Wurtsbaugh et al., 2019). In Lake Erie, phosphorus limits primary production early in the growing season, but N controls cyanobacteria blooms in late summer (Chaffin et al., 2013). Atmospheric deposition of N has also increased eutrophication in some lakes (Fenn et al., 2003; Bergström et al., 2006).

In addition to promoting eutrophication, N also can contribute to the toxicity of cyanobacterial blooms (HABs) in lakes (Davis et al., 2015; Gobler et al., 2016). This condition generally occurs in eutrophic and hypereutrophic lakes (Bogard et al., 2020; Hoffman et al., 2022), but is increasingly observed in non-eutrophic lakes including oligotrophic lakes that are strongly phosphorus limited with relatively high N concentrations (Townhill et al., 2018; Trainer et al., 2020; Howarth, 2022). Microcystin and many other toxins produced by cyanobacteria are highly enriched in N. Higher concentrations of N in the water column of lakes promote greater production of toxins (Dolman et al., 2012; Monchamp et al., 2014; Baker et al., 2018; Chaffin et al., 2018; Bogard et al., 2020; Hoffman et al., 2022). Across the Finger Lakes of New York State, the most toxic cyanobacterial blooms are found in the lakes with the highest water column N concentrations (NYS DEC 2018; Halfman et al., 2018), including oligotrophic and mesotrophic lakes that are strongly phosphorus limited (Roberts and Howarth, 2006). The production of toxins by cyanobacteria in lakes poses a major public health threat not only to those who drink water from lakes experiencing blooms but also from contact recreational activities. Exposure can also occur via the atmosphere due to the release of volatile toxins in areas near lakes and coastal waters experiencing harmful algal bloom events (U.S. EPA, 2023). In addition, higher production of toxins may have a positive feedback by increasing the size and duration of cyanobacteria blooms associated with reductions in grazing by zooplankton and benthic filter feeders (Howarth, 2022).

1.3 Coastal effects

Elevated N inputs is one of the greatest threats to the ecological integrity of coastal ecosystems, often causing eutrophication (the excessive production of algae and cyanobacteria), which can lead to hypoxic and anoxic waters (“dead zones”), degradation of habitat quality including loss of seagrasses and freshwater submerged macrophytes, fish and shellfish kills, decreased biodiversity, and increased harmful algal blooms (Vitousek et al., 1997; NRC, 2000; Bricker et al., 2008; Paerl and Barnard, 2020). The role of atmospherically derived N as a driver of eutrophication in aquatic ecosystems has long been recognized (Paerl, 1995, 2002; Howarth et al., 1996). A significant amount of the N entering aquatic ecosystems often is associated with atmospheric deposition, including both direct deposition to the surfaces of the water bodies and deposition onto their watersheds with subsequent export to the downstream wetlands, rivers lakes and estuaries. Estimates of the relative contribution of atmospheric N deposition to the total N load to estuaries range from a few percent to greater than 60% with values typically ranging from ~20 to over 40% depending on the area of the estuary relative to the area of the watershed and watershed development (Jaworski et al., 1997; Paerl et al., 2002; Boyer et al., 2002;

Castro and Driscoll, 2002; Castro et al., 2003; Whitall et al., 2003; Alexander et al., 2001; Burns et al., 2021).

In most of the estuaries and coastal marine waters of the United States, N is the primary element limiting rates of primary production, and increased loading of N to these ecosystems is the major cause of eutrophication (Howarth, 1988; Nixon, 1995; Vitousek & Howarth, 1991; NRC, 2000; Howarth & Marino, 2006; Paerl and Piehler, 2008; Howarth et al., 2021). Two-thirds of the estuaries in the country are moderately to severely degraded, and N is the single largest driver leading to degradation (NRC, 2000; Bricker et al., 2008). Notable examples of estuaries and coastal marine ecosystems degraded from excessive N inputs where the EPA is currently working with state and local stakeholders to reduce N inputs include Chesapeake Bay (MD and VA), Long Island Sound (NY and CT), Tampa Bay (FL), Barnegat Bay (NJ), Great Bay (NH and ME), Waquoit Bay (MA), and the “dead zone” in the Northern Gulf of Mexico near the plume of the Mississippi River (NRC, 2000; Rabalais et al., 2002; Bricker et al., 2008). One of the reasons that N is such an important driver of eutrophication in coastal ecosystems along the East and Gulf coasts of the United States is that deep-ocean sources provide a large input of phosphorus relative to N for these systems, making phosphorus limitation far less likely than in lakes (Howarth et al., 2021).

The downstream export of N to aquatic ecosystems from their watersheds is related to N inputs to the landscape from human activities. For large watersheds, these N inputs include fertilizer application, N fixation by agricultural crops, the net transport of N into or out of the watershed in human and animal food and feeds, and the atmospheric deposition of oxidized N (Howarth et al., 1996, 2006, 2012; Boyer and Howarth, 2008; Boyer et al., 2006; Hong et al., 2011, 2013; Goyette et al., 2016). The net sum of these inputs is called Net Anthropogenic Nitrogen Inputs (NANI). Note that atmospheric deposition of reduced N (e.g., ammonia, ammonium) is not included in this calculation, since it is assumed that most of the reduced N deposited was emitted to the atmosphere from the large watershed of interest and therefore is recycled within the watershed rather than a new input of N (Howarth et al., 2006, 2012; Hong et al., 2011, 2013). For smaller watersheds, however, atmospheric deposition of reduced N may represent a new input to the watershed. On average, approximately 25% of NANI is exported downstream to aquatic ecosystems, and importantly, this value applies equally to deposition of N and other N sources (Hong et al., 2011, 2013; Howarth et al., 2012). This rule of thumb can therefore be used to characterize the importance of atmospheric deposition as a source of the N inputs from watersheds to coastal marine ecosystems. Oxidized nitrogen deposition (NO_y) makes up 39% of total inputs on average for the watersheds in the northeastern United States (567 kg/km²-yr divided by the sum of 567, 310, 47, and 524 g/km²-yr) and 18% of the total net inputs for the Mississippi River basin, treating the export of N in food and feeds as a negative term (516 kg/km²-yr divided by the sum of 516, 2179, and 1,520 minus 1,421 g/km²-yr) (Figure 1). Note that the direct deposition of N onto the surface waters of aquatic ecosystems is an additional contribution of N to these ecosystems.

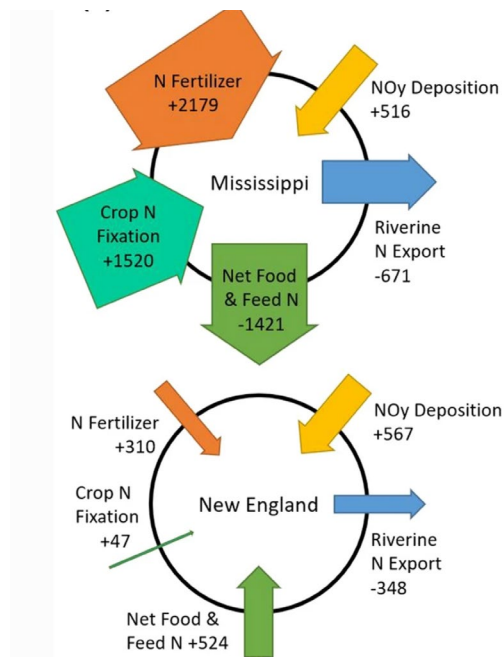


Figure 1 Comparison of inputs for net anthropogenic nitrogen inputs (NANI) in New England (lower) and Mississippi River basin (upper) in kg N/km²-yr. Also shown are exports of N in water flowing to coastal ecosystems based on 24% of NANI (after Howarth et al., 2021).

1.4 The Contribution of Atmospheric N Deposition to Total Maximum Daily Loads of Estuaries

There are widespread efforts across the U.S. to reduce N loads to estuaries through a TMDL approach, which is required by the Clean Water Act when a water body does not meet state water quality standards and its use is impaired. Atmospheric N deposition as a component of the total N load has been frequently targeted for reductions to meet TMDL standards. The CASAC provides examples of the role of atmospheric N deposition as a source of N to four estuaries along the U.S. Atlantic coast.

- Chesapeake Bay** has experienced significant impacts from eutrophication, including hypoxia of benthic habitat driven by excess nutrient loads, and efforts dating back to 1987 have targeted reductions in N, phosphorus, and sediment loads (NRC, 2000; Boesch et al., 2001). The Chesapeake Bay was the first estuary in the nation to target an explicit atmospheric deposition reduction goal as part of the 2010 TMDL allocation for N (Linker et al., 2013). The explicit allocation for direct atmospheric N deposition to the Chesapeake Bay and its tidal waters is 6.1 kg N/ha-yr by 2020 and is assigned to the EPA to evaluate in two-year increments. This allocation represents a 34% reduction from the 1985 atmospheric N deposition load and a 10% reduction from the 2010 load. Additionally, indirect atmospheric N deposition to the non-tidal portion of the Chesapeake Bay watershed is considered a reference allocation assigned to the individual states. Reductions in these indirect N loads are tracked by the Chesapeake Bay Program Airshed model and credited to states as appropriate, which reduces the required non-point source load reductions from manure and fertilizer that each state must meet as part of their TMDL requirements. A recent analysis indicates that annual atmospheric N deposition to the Chesapeake Bay watershed had declined to about 8 kg N/ha-yr as of 2021 suggesting that the TMDL goal had not yet been achieved, but a future projection suggests that the target load will be approached by mid-century (Burns et al., 2021). Recent analyses and model simulations indicate that total N loads to the bay are declining accompanied by increases in dissolved oxygen

concentrations. Declines in atmospheric N deposition have been identified as an important contributor to observed recovery patterns (Ator et al., 2019; Frankel et al., 2022; Zahran et al., 2022).

- **Waquoit Bay** and adjacent embayments on Cape Cod have experienced loss of eelgrass beds, decreased dissolved oxygen, periodic algal blooms and other deleterious ecological effects due to excess N loads to the bay (Commonwealth of Massachusetts, 2020). Atmospheric N deposition contributed about 35% of the total N loads to this estuarine system during 1992-94. A TMDL requirement was established in the Waquoit Bay system in 2013 to reduce N loads from multiple sources, but atmospheric N deposition was not considered a controllable source and therefore was not targeted for load reductions. Nevertheless, atmospheric N deposition declined by about 50% during 1990 – 2014, which compensated for similar magnitude increases in N loads resulting from urban land development during this period (Valiela et al., 2016). Atmospheric N deposition in 2017 was less than 5 kg N/ha-yr (Lloret et al., 2022) and has likely declined further since that date. These results highlight the benefit of decreasing atmospheric N deposition to estuarine ecosystems even when managerial controls available to local officials are deemed to have little influence on these loads (Valiela et al., 2016).
- Since the early 1990s, the **Tampa Bay** Estuary Program has had a principal objective to restore lost seagrass habitat to conditions assumed to have been present in the 1950s (<https://tbep.org/about-tbep/>). A nutrient management action plan establishes targets for N loads to the bay that uses a paradigm based on chlorophyll-a and light attenuation. Targets for direct atmospheric N deposition to the bay surface were established by Zarbock et al. (1994) based on a 1985-91 average value of 11.8 kg N/ha-yr. For the most recent 5-year period (2017-21) for which data are available, atmospheric N deposition has declined by more than 50% to 5.7 kg N/ha-yr (<https://tbep.org/tampa-bay-nitrogen-loads/>). This decline in atmospheric N deposition exceeds the management targets for the bay and coincides with declines in loads from other N sources. Water quality, chlorophyll-a, and light attenuation are improving in the bay. Atmospheric N deposition provided about 57% of the total N load to Tampa Bay in 2002, 17% from direct atmospheric deposition and 40% from indirect transport from the watershed (Poor et al., 2013). Direct atmospheric N deposition has declined to about 15% of total N loads to the bay as of 2017-21. There is not a specific indirect N load target for atmospheric N deposition, and the atmospheric contribution is not officially tracked by the nutrient management program. However, in 2003-07, non-point source loads were credited with a 32% decrease from atmospheric N deposition proportional to the decline for direct atmospheric N deposition to the bay surface, which lowered the non-point source requirements from other N sources proportionately.
- **The Neuse River Estuary**, NC, has suffered from N-driven harmful algal blooms that have been responsible for major fish kills, enhanced bottom water hypoxia and degradation of benthic and planktonic habitats (Paerl et al., 1995; Pzerl and Piehler, 2008). As a result, a N-driven TMDL was initiated in 1999 (<https://www.deq.nc.gov/water-quality/planning/bpu/neuse/neusetmdl1999phasei-0/download>). The TMDL mandates a 30% reduction in total N inputs to help stem further eutrophication and ecosystem degradation. Whitall et al. (2003) estimated that on average, 29% of total N loading to the estuary was due to combined indirect and direct atmospheric N deposition. Atmospheric N deposition of 9.8 kg N/ha-yr was estimated for the impaired area of the estuary during 1991-95 and targeted for a 30% reduction as for other N sources ([13](https://www.deq.nc.gov/water-</div><div data-bbox=)

quality/planning/bpu/neuse/neusetmdl1999phasei-0/download). A specific rule to promote targeted reductions in atmospheric N deposition was not established for the Neuse River Estuary, but it was noted in 1999 that atmospheric deposition was expected to decline significantly in the region by 2003 due to implementation of the Clean Air Act Amendments. An analysis with the Community Multiscale Air Quality (CMAQ) atmospheric chemistry model revealed that atmospheric N deposition had declined by 38% in the Little River tributary to the Neuse Estuary between 2000 and 2010 (Gabriel et al., 2014). A second tributary, the Nahunta Watershed, showed a lower decline of 19% because of a higher proportion of concentrated animal feedlot operations, which resulted in increased reduced atmospheric N deposition during 2000 to 2010. Model projections to the year 2020 indicated an expected decline of 51% in the Little River and 22% in the Nahunta. Recovery in the Neuse River Estuary (Gabriel et al., 2014) has been uneven with widespread decreases in nitrate concentrations but increases in ammonium and organic N concentrations (Lebo et al., 2011; Gabriel et al., 2018). As of 2009 and 2018, the North Carolina state standard for chlorophyll-a (40 µg/L) was still in exceedance in the Neuse River Estuary (Keith, 2014; Paerl et al., 2018).

Atmospheric N deposition provides a significant contribution to total N loads to U.S. estuaries that can range from a few percent to more than two-thirds. Atmospheric N is deposited directly to estuarine surfaces and is also supplied indirectly through transport by surface waters and groundwater. The four examples of estuaries along the U.S. Atlantic coast that are described here highlight widespread recognition of the importance of atmospheric N deposition as a source of ecosystem impairment where TMDL requirements have been required by the EPA. Approaches have varied from treating atmospheric N deposition as an uncontrollable source or a not specifically targeted source (Waquoit Bay, Neuse River Estuary) to establishment of a specific target load for direct atmospheric deposition (Chesapeake Bay, Tampa Bay). Indirect atmospheric N deposition has generally not been specifically targeted for reductions as part of the TMDL process, but decreased loads have been applied as a credit that lowers needed reductions in other non-point N sources such as in Chesapeake Bay and Tampa Bay. In general, atmospheric N deposition has declined from values of 10 kg N/ha-yr or more in the 1990s to values of about 5-8 kg N/ha-yr along the eastern U.S. coast in recent years. These declines have benefited efforts to reduce estuarine ecosystem impairments across the Atlantic and Gulf of Mexico coasts. In some cases, the decline in atmospheric N deposition has been described as one of the dominant contributors to ecosystem recovery, and even when little recovery has been evident, decreases in atmospheric N deposition have compensated for increases in other N sources. In conclusion, declines in atmospheric N deposition that have resulted from implementation of the Clean Air Act have served to benefit efforts to minimize ecosystem impairment to U.S. coastal estuaries. Despite these improvements, the TMDL goals of these estuaries have not yet been achieved. Additional future reductions in atmospheric N deposition would help facilitate the recovery of these impaired estuaries.

2. Acidification

2.1. Chapter 5 Analysis Decisions for Acidification

In addition to concerns about the inadequate treatment of N in the PA, the CASAC has concerns about the consideration of acidification effects. The CASAC requests that the EPA address the following questions on acidification in the PA. The PA assumes that atmospheric S deposition is the dominant driver of acidifications effects. But does using S deposition as the indicator for freshwater acidification protect ecosystems from nitrate driven episodic acidification? The PA should address the effects of N+S deposition on freshwater acidification and critically evaluate the assumption that acidification effects

only due to atmospheric S deposition (and not N deposition) are adequately protective of freshwater ecosystems.

2.2. Indicators of acidification and limitations (ANC, BC:Al, soil health, C/N)

The ratio of base cations (Ca+Mg+Na+K) to aluminum (BC:Al) in soils has been a commonly applied index to reflect risk to tree growth and survival from the acidifying effects of atmospheric deposition (Sverdrup and Warfvinge, 1993; Cronan and Grigal, 1995). While the abundance and availability of base cations and aluminum in soils have proven to be important in governing tree health, the application of specific threshold values of the BC:Al ratio to indicate risk to tree health has proven to be problematic (Falkengren-Grerup et al., 1995; Tyler and Falkengren-Grerup, 1998). Early studies of the effects of BC:Al as reviewed by Sverdrup and Warfvinge (1993) and Cronan and Grigal (1995) were largely based on soil solution concentrations derived from greenhouse experiments. Thresholds of harm to tree health originating from these early studies did not translate well to *in-situ* observations in which a variety of factors such as the presence of soil organic matter can affect the speciation and availability of aluminum. Recent investigations have favored exchangeable calcium or soil base saturation (BS, sum of BC/soil exchange capacity expressed as percent) as more accurate metrics of risk to tree species sensitive to acidification such as red spruce and sugar maple, and a threshold base saturation value in the range of 12 to 16% has proven useful in tree health assessment (Sullivan et al., 2013; Lawrence et al., 2017). Use of these metrics reflects the importance of exchangeable calcium in soil as a nutrient that can limit tree growth when depleted from soils as well as its role as a stress indicator (Schaberg et al., 2001). Furthermore, soil solution chemistry is not widely measured in many catchments limiting the availability of extensive BC:Al data sets available for analysis. Assessment of thresholds of harm to forested ecosystems using soil base saturation reflects system *capacity*, whereas a preference to BC:Al in soil solution reflects system *intensity* (Holmberg et al., 2001). The BC:Al ratio in soil solution continues to have value as an indicator, however, negative effects on tree health are not always observed when values are <1 (Hansen et al., 2007) as originally proposed by Cronan and Grigal (1995). Some investigations have evaluated thresholds of harm to terrestrial ecosystems by applying BC:Al in soil solution as well as soil base saturation, and results are often comparable suggesting that either approach is viable in critical loads assessments (Sullivan et al., 2011). These studies also underscore the critical importance of clearly stating whether these metrics of soil cation status are referring to soils or soil solutions, a common complicating factor in the literature as well as in the discussion in the PA. The CASAC recommends that the EPA deemphasize the use of BC:Al as a terrestrial acidification indicator in the PA and emphasize soil base saturation because it is a more robust and commonly accepted indicator.

The PA also fails to adequately discuss soil carbon to N ratios (C/N) as an important indicator of ecosystem N status. Earlier literature has established C/N as an effective indicator of the degree of N enrichment in terrestrial ecosystems such as forests (Aber et al., 2003) and moorlands (Evans et al., 2006), often providing soil thresholds for accelerated nitrification and nitrate leaching among other ecosystem processes. Recent literature continues to utilize C/N as an indicator of ecosystem N status (e.g., Cheng et al., 2019, 2020; Newcomer et al., 2021; Tahovská et al., 2020; Yu et al., 2018) and this body of science warrants further discussion in these chapters. The CASAC recommends that the EPA introduce and discuss soil C/N as an indicator of ecosystem N status in the PA.

2.3. Steady State vs Dynamic CL models

Much of the analysis on aquatic and terrestrial effects of acidification presented in Chapter 5 is based on steady-state modeling. The CASAC requests that the text of the PA in Chapter 5 be expanded to include

a description of the assumptions associated with steady state modeling and the limitations of the application of such models (e.g., see McDonnell et al., 2023). Steady-state models are time independent. Acidification steady-state models assume that soil pools remain constant with respect to the specified deposition loading and simulated surface water chemistry and provide no perspective about the rate or pathway of chemical recovery of ecosystems. Steady state models are a valuable tool with few required inputs and parameters and therefore can be widely applied. The analysis conducted by EPA on steady-state critical loads is impressive. However, it is important to note that real ecosystems are never at steady-state. Chemical pools and fluxes change over time and these changes influence ecosystem sensitivity to ongoing acid deposition and the time frame of recovery, both critical policy issues. The CASAC asks that in revision of the PA, the EPA clearly indicate the assumptions and limitations associated with steady-state model calculations. The PA mentions but does not elaborate on dynamic acidification models. Dynamic models are also an important tool. Dynamic models can be used in tandem with steady state models to provide important perspective on the time-dependence of acidification or recovery and the processes that are driving ecosystem response. There have been important efforts in dynamic modeling in recent years that could bolster information on effects on soil nutrient concentrations and pools and the time frame for watershed ecosystem recovery (see McDonnell et al., 2021, 2023; Shao et al., 2020 2021; Fakhraei et al., 2016, 2017). The EPA needs to include a discussion of dynamic modeling with examples of dynamic modeling results in Chapter 5 to provide important context to the steady-state critical load modeling results presented.

3. Climate/legacy deposition implications

Two areas of consideration regarding the uncertainty in our current scientific knowledge are understated in the draft PA. The first is, as stated in the PA (5-51), that the "...extent of the influence of historical deposition...on the associations reported in these studies with metrics quantifying more recent deposition is generally not known." Past research conducted during periods of historically higher rates of N and S deposition adds uncertainty to what we 'know' about how well that research applies to the future period of ecosystem recovery. For example, research shows that geographical differences in the eastern US, notably between glaciated and non-glaciated regions, alters the rate of ecosystem recovery and the influence of historical air quality varies in current and future ecosystem responses to ambient deposition (Burns et al., 2020; Harmon et al., 2021; Rice et al., 2014). Similarly, recent literature (e.g., Gilliam et al., 2023; Newcomer et al., 2021; Patel et al., 2020) highlights the complexities and characteristic hysteresis in ecosystem recovery from elevated atmospheric N and S deposition in forests that is not included in the PA. These patterns underscore the added uncertainty with which past research conducted during periods of historically high air pollutant exposures can be applied to future ecosystem responses that is not adequately developed in the draft PA. The CASAC requests that the EPA improve its analysis of uncertainty pertaining to the assumption that past observations can be used with confidence to project future ecosystem response to effects of S and N.

The second area of consideration is that the PA makes only passing reference to the reality that knowledge of N, S and fine PM impacts on ecosystems is derived from past research, and the uncertainty around these impacts in the future is influenced by the accelerating changes in the climate system (IPCC, 2023), including warming, intensification of hydrologic cycles, and increasing atmospheric CO₂ among others. The PA refers to climate mostly in the context of historical climate as a factor considered in several studies referenced. There is an increasing body of literature on the intersection of a changing climate and N, S, and PM secondary effects. Examples of this body of research include the influences of increasing atmospheric CO₂ and N on tree biomass (Wason and Dovciak, 2017; Wang and Wang, 2021), forest growth responses to acid deposition recovery and climate

(Kosiba et al., 2018; LeDuc et al., 2022; Clark et al., 2023), temperature and precipitation effects on weathering of base cations (Kopáček et al., 2017), shifting climate versus air quality effects on the terrestrial carbon cycle and dissolved organic carbon export to surface waters (e.g., de Wit et al., 2021), the interactions of changing water chemistry and climate on mountain ponds in the Northeastern U.S. as sentinels of change (Nelson et al., 2021), and the mitigating effect of acid rain recovery on climate change impacts on sensitive fish populations (Warren et al., 2017). This emerging body of research implies that the uncertainty associated with relying on past research to predict future welfare effects is increasing, and that it is essential to incorporate the shifting climate reality into decision-making. The PA can more clearly convey that the established science of past decades may not translate unaltered into the future. In addition, as noted in the CASAC letter to the Administrator on May 5, 2020, in review of the ISA Chapter 1.12, “The CASAC also finds that there is a need for clarification of the scope of what is considered to be “climate” (meteorological factors over a specific time period), in contrast to long-term climate change, or associated changes to CO₂ concentrations and impacts on biogeochemistry.” These are distinctions that are of increasing importance in the assessment of the science and its relevance to risk in the PA.

4. General comment

A good general discussion of public welfare effects is presented in section 4.3 of the PA. In this section it is rightly noted that the Administrator ultimately decides whether the effects of NO_x, SO_x and PM rise to the level of adverse effects, but more could be said to help in this decision. The last row in Figure 4.3 can be much more assertive about describing the public welfare effects of atmospheric N and S deposition. These are not “possible”; but rather there is considerable confidence that these effects occur. Moreover, they fall within the definition of public welfare effects. These effects span direct human usage and enjoyment of the environment and the fact that people care about protection of biodiversity and healthy habitats even if they never visit these places. Bringing forward some of the discussion of ecosystem services from the ISA may be helpful here. What is needed in the PA is to make clear why the effects described in the previous sections are important. More specific summary descriptions would help emphasize the results discussed in the previous sections. For example, indicate that acidification of streams and lakes has resulted in loss of sensitive species of fish and other biota, including fish species popular with anglers. Further, effects don’t have to be fully quantified in order to be confident that they are happening. For example, it may not be possible to indicate the specific amount that atmospheric N deposition is contributing to eutrophication in an estuary or the toxicity of HABs in a lake, but there is confidence that it is contributing and therefore causing associated public welfare effects.

Chapter 6 – Relationships of Deposition to Air Quality Metrics

To what extent does the Panel find the investigation of relationships between air concentration and deposition to be technically sound and clearly communicated? What are the CASAC views on the consideration of key limitations and associated uncertainties?

In Chapter 6, an extensive evaluation of the relationships between air quality metrics and deposition of N and S to ecosystems is presented. The evaluation is conducted for Class I areas and for the entire U.S. In the Class I areas analysis, EPA presents a series of scatterplot matrices showing relationships between measured PM_{2.5} constituents from various monitoring networks and measured wet N and S deposition and between measured PM_{2.5} constituents and modeled values of N and S deposition from the CMAQ and Total Deposition (TDEP) models. Most of the figures show expected relationships and provide

useful information. However, the number of figures is overwhelming, and the interpretation provided is brief. The CASAC recommends that the EPA provide more elaboration on the most useful relationships. Separation of reduced from oxidized N deposition, where possible, would also be elucidating. In addition, several figures are characterized by non-random patterns (bifurcations and trifurcations), especially for the N species. It is possible that the patterns are caused by a subset of sites in a particular geographic area, or there may be other plausible explanations. Because a significant portion of the data do not conform to the expected relationships, additional explanation of these anomalies by the EPA is needed to make this chapter more useful and interpretable. The scales and units of these figures should be clarified (e.g., “Total Deposition” and “IMPROVE N+S” in Figure 6-10; “Total Deposition” and “CASTNET N+S” in Figure 6-11). Often, there is a lack of consistency in how the figures are described. Given the number of figures, the CASAC recommends that they be harmonized. Harmonizing their presentation is necessary to enable the reader to make accurate comparisons and judgments about these important relationships.

In the national scale sites-of-influence analysis (Sec. 6.2.2), the EPA uses the National Oceanic and Atmospheric Administration (NOAA) Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model to establish quantitative relationships between ambient pollutant concentrations at upwind source sites (or “sites of influence”) and deposition of N and S at downwind sites. As noted in its overarching comments on deposition-based standards, the CASAC recognizes the fundamental difficulties of finding quantitative relationships between air quality concentrations and deposition. The CASAC further acknowledges that the problem of using atmospheric measurements to predict deposition has not been adequately researched or documented in the peer-reviewed literature. This shortcoming required the EPA to develop the Ecoregion Air Quality Metric (EAQM) methodology in-house. The EAQM results presented in Chapter 6 are used in Chapter 7 to support the secondary NAAQS recommendations.

The CASAC does not have consensus on its response to the technical approach taken. Most CASAC members are comfortable with the EAQM modeling approach, given the prior use of non-peer reviewed analyses in REAs, but indicate that this approach and methodology require much more detailed explanation in the text. Multiple CASAC members believe that the use of an approach that has not been peer reviewed is not appropriate for a standard setting basis and one panel member does not believe that it is the responsibility of the CASAC to provide such peer review. That said, some CASAC members find the EAQM results useful since there are limited analyses available that compare SO₂, NO₂, and PM_{2.5} design values to S and N deposition values. However, other CASAC members find that the EAQM results are not scientifically sound and should not be used to inform the secondary standard recommendations for SO₂, NO₂, and PM_{2.5}. This concern is especially evident when examining more recent design values (2014-2016 and 2018-2020) compared to older design values (2001-2003, 2006-2008, and 2010-2012), and consideration of the Western U.S. compared to the Eastern U.S. ecoregions. Please see individual panel member comments for details.

The CASAC finds that the description of the EAQM method in Section 6.2.2 lacks adequate detail. Although readers are referred to Appendix 6A., the appendix contains only one reference to HYSPLIT and very little discussion of the methodology. Many of the parameters and assumptions need to be explained and justified. These include how the meteorology, trajectory characteristics (e.g., number, height, start time, duration), number of “hits”, and contribution thresholds were determined. In addition, the CASAC recommends that the EPA conduct sensitivity runs using different assumptions and parameter values to quantify the uncertainty associated with this approach.

Assuming the EPA continues to use the EAQM approach, the CASAC offers the following detailed comments:

- Appendix 6A needs to include the equations that were used to calculate EAQM-weighted concentrations, EAQM-max concentrations, and the median S and N deposition values. The Appendix needs to explain why median deposition values were used instead of mean deposition values. Also, the EPA should provide a simplified example showing how the equations are applied to generate the final EAQM values.
- In Figure 6A-1, clarify the purpose of the grey circles in the middle of the ecoregion since every trajectory originating from these monitoring locations will pass through large portions of the ecoregion and contribute to the deposition in that ecoregion. The appendix should provide a more detailed explanation of how the number of “hits” and percentage of hits were determined for each monitor. Again, the EPA should provide a simplified example showing how these calculations were performed.
- The EPA needs to elaborate more on why 2016 is selected as the study year and demonstrate that this single year of meteorology is representative of other recent years. The CASAC recommends that the EPA run at least three years of meteorology to match number of years used to calculate design values. At a minimum, the EPA needs to create annual wind roses for 2016 and compare these to 3-year average wind roses to justify that this one year of meteorology is representative.
- The EPA should clarify how many trajectories are released from each monitor location each day. Also, the start time for each trajectory should be documented.
- The EPA should justify why 48-hour trajectories were used. Since secondary species and PM have lifetimes of 2-10 days and can be transported long distances across multiple states, it might be more appropriate to use 72-hour or 96-hour trajectories.
- The EPA should justify why a 1% contribution threshold is appropriate. The CASAC believes that the contribution threshold should be lower (e.g., 0.5%, 0.1%, etc.) to increase the number of air quality monitors included in the analysis for each ecoregion.
- The EAQM approach based on HYSPLIT does not account for chemical transformations and as a result, would not address situations where emissions (e.g., SO_x or NO_x) in one location could reduce deposition at a location downwind by promoting secondary PM formation that is transported further than the target area. In general, chemistry and thermodynamics should not be neglected when considering air/deposition relationships.
- The use of nearby SO₂ and NO₂ monitors to evaluate S and N deposition inside the ecoregion may not always give the best reflection of deposition.
- The current ambient monitoring network for SO₂ may not capture the impacts from many of the large SO₂ industrial emission sources. Also, the current NO₂ ambient monitoring network is not designed to capture the impacts from large NO₂ industrial emission sources and is very scarce in many parts of the county. In some cases, the SO₂ and NO₂ ambient monitoring network may not be adequate to represent the spatial gradients in the ecoregion.
- Due to the seasonal nature of nitrate formation and NH₃ sources, reactive N transport and deposition will be seasonal. The EAQM values are based on transport patterns over the entire year and may not be indicative of the transport patterns during the “deposition season.”
- The EPA’s EAQM approach does not account for the significant contributions of nitric acid wet and dry deposition to N deposition.
- Total PM_{2.5} may be a poor indicator of sulfate (SO₄ PM_{2.5}), nitrate (NO₃ PM_{2.5}), and ammonium (NH₄ PM_{2.5}). Based on the 2019-2021 speciated PM_{2.5} data in Figure 2-26, only 20-40% of total PM_{2.5} is ammonium sulfate and ammonium nitrate. Moreover, in this figure, OC is presented rather than OM. If one converts the OC to OM (by multiplying OC*1.4) and includes the

unidentified PM_{2.5} mass (typically OM), then the percent contributions from ammonium sulfate and ammonium nitrate decrease to 15-30%. Also, these percentages are even smaller on days that exceed the current 24-hour standard since the additional PM_{2.5} mass on those days are typically organic carbon and elemental carbon from wildfires and prescribed fires, or crustal material from dust events.

- EPA should perform sensitivity runs using different assumptions to evaluate the robustness of the relationships between deposition and EAQM values with regards to these assumptions and help quantify the uncertainty associated with this approach. The sensitivity runs should include (1) the numbers of years of meteorology, (2) number of trajectories released each day, (3) the start time for each trajectory, (4) the starting trajectory height, (5) the duration of the trajectories, (6) the contribution threshold, and (7) seasonality of deposition.

If this approach is continued, updates to the EAQM analysis based on CASAC's advice should be included in the Second Draft PA.

Appendix 6A consists mostly of tables of results, but this format makes it very difficult to determine similarities and differences in the temporal or geographic output. The EPA should expand and document the EAQM model description in both Chapter 6 and Appendix 6A and provide the EAQM output in a format that facilitates interpretation of the results – i.e., maps or figures to allow spatial and temporal comparisons. In addition to the current single figure showing a single ecoregion for a single pollutant (Figure 6A-1), the document should include maps of all the ecoregions and all the pollutants (SO₂, NO₂, and PM_{2.5}). Tables similar to Tables 6A-1 through 6A-4 should be included for all combinations of ambient air pollutant concentrations and deposition values that were examined. In addition, these tables should include the number of SO₂, NO₂, and PM_{2.5} “monitoring sites of influence” that are included in the calculations for each ecoregion.

In particular, the very strong correlation ($r = 0.98$) between N deposition and the EAQM weighted annual average PM_{2.5} in the original Figure 6-25 is unexpected and it is unclear what factors could account for it. Comparing Figure 2-38 (three-year average of the total deposition of N (kg N/ha) across the 2019-2021 period) to Figure 2-27 (annual PM_{2.5} design values for the 2019-2021 period), the correlation between N deposition and annual PM_{2.5} design values is very poor compared to the very strong correlation shown in the original Figure 6-25. This situation may call into question the validity of the EAQM approach. Because this is one of the relationships (among others) that is supportive of the proposed use of a PM_{2.5} concentration standard to control deposition, it warrants more explanation.

On July 24, the EPA released a memo titled “Correction to Errors in Six Figures of Chapter 6 of the Policy Assessment for the Review of the Secondary National Ambient Air Quality Standards for Oxides of Nitrogen, Oxides of Sulfur and Particulate Matter, External Review Draft.” The EPA followed this up on July 27 with a memo that also provides a table of correlation coefficients for correlations of deposition estimates with various air quality metrics as well as attachments with the corrected figures embedded in the corrected text of the PA. These memos correct Figures 6-20, 6-21, 6-25, 6-26, 6-28, 6-29. The corrected figures address the concerns raised with the original Figure 6-25 in that the actual correlation between the EAQM weighted annual average PM_{2.5} and N deposition is modest ($r = 0.52$), which is more consistent with the contents of Chapter 2. Further, the relationship between N deposition and NO₂ annual average concentrations is shown, in the corrected versions, to be non-existent across all ecoregions, which is again consistent with the contents of Chapter 2. However, the corrections and updated text do not address remaining questions that the CASAC has about the data in Chapter 6.

Additionally, the corrected figures (especially Figures 6-20, 6-21, 6-25, and 6-26) show highly pronounced bifurcation / clustering in the plotted relationships. This bifurcation is not addressed in the associated text within Chapter 6 of the PA or in EPA's corrections. This situation underscores the CASAC's request for additional analyses of the spatial distribution of these relationships, or their dependence on specific chemical forms (oxidized vs reduced) of N deposition. The CASAC recognizes that the memo communicating these corrections includes additional consideration of spatial differences, i.e., correlations in Eastern versus Western ecoregions. However, despite the grouping of Eastern versus Western ecoregions seeming to be a useful distinction in this analysis, this grouping is not mentioned anywhere within the corrected text to Chapter 6. Thus, without incorporation of this analysis into the PA itself, the CASAC is unable to comment on the utility of this distinction, as there is no discussion of standards being considered in a spatially heterogeneous fashion. The CASAC thus requests a revised analysis that explicitly addresses this topic. Further, it will be very useful for the CASAC's evaluation of the EPA's standard recommendations in Chapter 7 if the EPA could parse out statistical relationships in Figures 6-20, 6-21, 6-25, 6-26, 6-28, and 6-29 separately for all data and for the two most recent time periods. Given changes to ambient atmospheric chemical conditions, the latter may be most relevant.

Chapter 7 – Review of the Standards

What are the Panel views on the discussion of key considerations in sections 7.1 and 7.2 and preliminary conclusions? What are the CASAC views regarding the areas for additional research identified in Chapters 3 and 4? Are there additional areas that should be highlighted?

In Chapter 7, the EPA examines the scientific evidence reviewed as part of the 2020 ISA as well as air quality and exposure analyses to determine whether the current secondary standards for NO_x, SO_x, and PM provide adequate protection against associated welfare effects. The chapter concludes with a presentation of potential policy options to address air-related and deposition-related effects of N and S on ecosystems (Table 7-1).

The CASAC does not find the information on potential welfare effects of N and S in ambient air and in deposition reviewed in the ISA and PA to be adequately summarized (in part due to the literature being reviewed only until 2017), as noted in the Chapters 4 and 5 consensus responses. As discussed in more detail above and below, the CASAC urges the EPA to consider secondary standards based on deposition rather than exposure given the strong, decades-long evidence for deposition-related effects on ecosystems.

To protect public welfare against the effects of NO_x, SO_x, and PM, the CASAC believes that the secondary standard should be based on the strongest existing evidence for ecological effects of these pollutants. Several decades of research show that ecosystem effects are largely manifested through atmospheric deposition as presented in the 2020 ISA. The ISA reviews N and S deposition effects on six categories of ecosystem structure and function in terrestrial, wetland, freshwater, and estuarine ecosystems: (1) ecosystem productivity, (2) community biodiversity, (3) population/individual growth, (4) individual physiology, (5) soil/sediment chemistry and (6) surface water geochemistry via enrichment, eutrophication, sulfide toxicity, mercury methylation, and acidification. All the categories considered in the 2020 ISA review were determined to be causal.

In contrast, relating atmospheric deposition-based effects to ambient concentrations is fraught with difficulties. The CASAC did not reach consensus on the approach taken in the 2023 PA to establish

quantitative relationships between ambient pollutant concentrations at upwind source sites and deposition of N and S at downwind sites, but the CASAC reached consensus that the description and methods need improvement. Further, the results derived from this quantitative analysis showed generally that relationships between air quality metrics and deposition are not consistent across all ecoregions.

The CASAC thus recommends that direct atmospheric deposition standards be considered in future reviews. Direct atmospheric deposition of S and N pollutants is a clear and straightforward indicator of “the presence of these pollutants in the ambient air.” Indeed, since wet deposition reflects the removal of pollutants through a deep column of the atmosphere, it is a much better indicator than could be derived from surface air monitors of the presence of the deposited pollutants in the ambient air, and is also much more directly relevant to the ecological effects of these pollutants. The current PA and the supporting ISA both depend heavily on annual estimates of total wet and dry deposition of S and N species produced by the National Atmospheric Deposition Program (NADP) Total Deposition (TDep) Science Committee. The TDep approach, initially described by Schwede and Lear (2014), combines state of the art monitoring and modeling data to produce the consensus highest quality annual estimates of wet, dry and total deposition of various S and N (and other relevant) species, currently with 4 km spatial resolution over the coterminous U.S. This approach has been continually refined over the past decade, with improvements both applied to historical data and carefully described in annual TDep methods documentation files (NADP, 2023). The measurements, modeling, and methods for blending them and assessing their results are conducted, supported and refined by a wide range of scientists from states, academia, the EPA, multiple other federal agencies and Environment Canada. Results from a significant recent enhancement of TDep methods and results will be presented shortly for peer review by Beachly et al. (in preparation). Given that the objective of a secondary NO_x/SO_x/PM NAAQS indicator would be to represent annual deposition over broad spatial areas (ecoregions), the annual TDep estimates are uniquely and ideally suited for this application.

Traditionally, the EPA has relied exclusively on near-surface air quality measurements, with formal “Federal Reference Methods” developed by the EPA and delegated to the states, as the basis for determining compliance with primary and secondary NAAQS. Given the clear importance of deposition to ecological effects of S, N and PM, and the complexities and uncertainties associated with linking near-surface air quality measurements to (nearby) deposition, the TDep data provide a superior approach for assessing both temporal and spatial deposition of S and N. The broad, multi-agency support for NADP TDep calculations helps assure continued future scientific and logistical support for the future.

Nonetheless, the CASAC provides the following recommendations for alternative standards based on air concentrations that should be protective of direct and deposition-related effects. The EPA should incorporate these recommended alternative standards into the Second Draft PA. In addition, because of the cumulative effects of deposition on ecosystem structure and function, longer averaging times are better suited to address deposition-related effects of N and S than shorter ones. The revisions to the standards that the CASAC is putting forth for consideration are generally supported by the evidence reviewed in the ISA, in the draft PA, and in the July 24 and 27, 2023 follow-up memos. The recommendations below frequently refer to figures in Chapter 6 based on the ecoregion analysis conducted by the EPA using the EAQM model. Many of the correlations therein between ambient concentration metrics and S or N deposition are notably small, statistically. The CASAC notes, however, that in these figures, there are often identifiable thresholds of ambient concentrations below which deposition levels are of less concern, even when the direct correlations between concentrations and deposition at higher concentration values are minimal. Corroboration with additional evidence (such as analysis based on TDep) is referenced where available.

The CASAC finds that the EPA's justification for the potential alternative standards needs to be more clearly explained, with additional detail provided regarding the ecosystem effects of the proposed ranges. The CASAC recommends that as part of the presentation of each policy option, the PA refer the reader to the relevant supporting tables and figures (e.g., figures and tables in Chapters 2, 4, and 6). Finally, the CASAC welcomes the potential for a mass- or composition-based PM-based standard to reduce N and S deposition effects on ecosystems. While the CASAC provides recommendations on potential alternative standards based on the evidence presented in the PA, it requests that the EPA provide a more quantitative assessment of the data and indicate what additional amount of protection would be achieved at each recommended level.

The CASAC has consensus regarding the 3-hour SO₂ standard for direct effects. The current SO₂ secondary standard of 0.5 ppm (3-hour maximum not to be exceeded more than once per year) is adequate to protect against the direct effects of oxides of S on plants and lichens and should be retained without revision.

The CASAC also has consensus regarding the annual NO₂ standard for direct effects. The current annual secondary NO₂ standard of 53 ppb is adequate to protect against the direct welfare effects of N oxides in ambient air on plants and lichens and should be retained without revision.

All of the CASAC members,* except one, have consensus on the following recommendations regarding deposition-related effects:

Annual PM_{2.5} standard

Recommendation: The current secondary PM_{2.5} standard is inadequate to protect ecosystems against the deposition-related effects of N and S and a secondary annual PM_{2.5} standard in the range of 6-10 µg/m³ is recommended.

Justification: In remote areas, IMPROVE PM_{2.5} concentrations in the range of 2-8 µg/m³ for the periods 2014-2016 and 2017-2019 correspond with total S deposition levels <5 kg/ha-yr (Figure 6-12), with levels generally below 3 kg/ha-yr, and with total N deposition levels ≤10 kg/ha-yr (Figure 6-13). Ten kg/ha-yr is at the middle to upper end of the N critical load threshold for numerous species effects (e.g., richness) and ecosystem effects (e.g., tree growth) in U.S. forests grasslands, deserts, and shrublands (e.g., Pardo et al., 2011; Simkin et al., 2016) and thus 10 kg/ha-yr provides a good benchmark for assessing the deposition-related effects of NO₂ in ambient air. Tree survival and growth effects are widely documented at S deposition rates >5 kg/ha-yr (Pavlovic et al., 2023). Thus, total N deposition levels ≤10 kg/ha-yr and total S deposition rates ≤5 kg/ha-yr afford an adequate level of protection to several species and ecosystems across the U.S.

Higher (hotspots of) total N deposition (>15 kg/ha-yr) in California, the Midwest, and the East (Figure 2-38) correspond with areas where secondary annual PM_{2.5} design values for 2019-2021 are in the range of 6-12 µg/m³ (Figure 2-27). In the ecoregion analysis, which encompasses areas near sources, annual average PM_{2.5} design values in the range of 6-12 µg/m³ are also associated with total S deposition levels ≤5 kg/ha-yr (Figure 6-24). Total N deposition levels <10 kg/ha-yr

* This also includes all of the CASAC Panel members, except for the one CASAC member and one additional panel member.

are only achieved at EAQM weighted annual average PM_{2.5} values of <6 µg/m³ (Figure 6-25) and <8 µg/m³ for maximum annual average ecoregion PM_{2.5} (Figure 6-26).

The CASASC notes that the correlation between annual average PM_{2.5} and N deposition is 0.52 when the weighted average is considered. Correlations are minimal (0.03 for N deposition, 0.12 for N+S) when the contributing monitor with the maximum value for each ecoregion is considered. Correlation coefficients between annual average PM_{2.5} and S deposition are not presented in Attachment 1 in the July 27 memo.

Given the weaker relationships between PM_{2.5} and N and S deposition presented in the July 27, 2023 memo to the CASAC than presented in the PA, a more detailed evaluation of the recent relationships between PM_{2.5} N, PM_{2.5} S, PM_{2.5} N+S vs. N, S and N+S deposition is recommended.

A new total N PM_{2.5} indicator is recommended to achieve a better measure of total reactive N deposition, including ammonium deposition. A PM_{2.5} mass indicator may be adequate for now but may not be adequate in the future due to shifts in PM_{2.5} composition across space and over time as a result of changing wildfire and dust regimes, for example. Further, total PM_{2.5} mass underestimates the amount of N in PM_{2.5} due to evaporative nitrate losses on filter samples.

Annual NO₂ standard – Deposition-related effects

Recommendation: The current annual secondary NO₂ standard of 53 ppb does not provide sufficient protection against the deposition-related welfare effects of N and is therefore inadequate. The secondary annual NO₂ standard should be revised to <10-20 ppb.

Justification: At national scales, there are not strong correlations between average NO₂ concentrations and N deposition. However, focusing on the cluster of data with higher N deposition values, at a weighted annual average NO₂ concentration of <5 ppb (averaged over a 3-year period), total N deposition is ≤10 kg/ha-yr. At weighted annual average NO₂ concentration of <10 ppb there are ~12 exceptions after 2010 (Figure 6-20).

According to Attachment 1 of the July 27 EPA memo to the CASAC, when considering all ecoregions, there is no correlation between annual average NO₂ and N deposition. There appear to be stronger correlations when analyzing the data regionally, as explained in the July 27 memo. According to this memo, for the Western ecoregions, correlations are negligible, however they are low to moderate in the Eastern ecoregions. Still, within Eastern ecoregions the weighted average NO₂ value is more strongly correlated than when the contributing monitor with the maximum value for each ecoregion is considered.

Annual SO₂ standard – Deposition-related effects

Recommendation: In contrast to the direct welfare effects of SO₂ in ambient air, the current SO₂ standard does not afford adequate protection against the deposition-related effects of S on ecosystems. A new secondary annual SO₂ standard is recommended in the range of 10-15 ppb to preclude the possibility of returning to deleterious deposition values as observed associated with the emergence of high annual average SO₂ concentrations near industrial sources in 2019, 2020, and 2021 (Figure 2-25).

Justification: This level generally maintains S deposition at <5 kg/ha-yr for the periods 2014-2016 and 2018-2020 (Figures 6-17 and 6-18), affording protection to various tree and lichen species (Geiser et al., 2021; Pavlovic et al., 2023), and is the level at which 80%, 80%, and 70% of waterbodies per ecoregion were estimated to achieve ANC at or above 20, 30, and 50 µeq/L, respectively, in all ecoregion-time period combinations.

24-hour PM_{2.5} standard

Recommendation: The current 24-hour standard is not adequate to protect against short-term episodic events. A 24-hour PM_{2.5} secondary standard of 25 µg/m³ or a level of 20-25 deciviews based on a PM_{2.5} chemical light extinction indicator is recommended.

Justification: While the PA appropriately emphasizes ecological effects resulting from cumulative long-term deposition of S, N, and PM components, there is strong seasonal variability in emissions, transformation chemistry, stagnation, and transport. These can result in large seasonal variations in source/receptor relationships and in wet and dry deposition of different S and N species that are not adequately captured by long-term averages. Ecological sensitivities can also exhibit strong seasonal variabilities. Sensitive lichen species are especially dependent on deposition from fog or cloud water, from which S and N contributions can be highly episodic (and are typically not characterized in existing wet or dry deposition monitoring networks).

For these reasons, a separate 24-hour PM_{2.5} standard, or 24-hour standards based on PM_{2.5} species, would provide added protection against seasonal or episodic ecological effects. Visibility impairment, another important PM_{2.5} welfare effect is also most appropriately considered over shorter 24-hour or sub-daily averaging times. In announcing the most recent proposed revisions to the PM NAAQS, the Administrator solicited comments on “the appropriateness of a target level of protection for visibility below 30 dv and down as low as 25 dv, and of revising the level of the current secondary 24-hour PM_{2.5} standard to a level as low as 25 µg/m³. (Fed. Reg. 88, No. 18 / Friday, January 27, 2023 / Proposed Rules, p. 5662). Twenty-five deciviews is the level that is accepted by ~50% of the preference study participants that is documented in the NAAQS PM review. Thus, when considering protection of both sensitive ecosystems and visibility, the CASAC recommends a 24-hour PM_{2.5} secondary standard of 25 µg/m³ or a level of 20-25 deciviews based on a PM_{2.5} chemical light extinction.

However, one CASAC member finds that the historical TDep values demonstrate that the current primary NAAQS for SO₂, NO₂, and annual PM_{2.5} provide adequate protection against “long-term” annual S and N deposition-related effects; therefore, the secondary NAAQS for SO₂, NO₂, and annual PM_{2.5} should be set equal to the current primary NAAQS for SO₂, NO₂, and annual PM_{2.5}. Please see Appendix A for details on the dissenting viewpoint.

Network Funding

The CASAC wishes to emphasize that virtually all the research in this PA depends heavily on data from our various national monitoring networks: National Air Monitoring Stations (NAMS)/State and Local Air Monitoring Stations (SLAMS), Chemical Speciation Network (CSN), Interagency Monitoring of

Protected Visual Environments (IMPROVE), Clean Air Status and Trends Network (CASTNET), National Atmospheric Deposition Program (NADP), and more. Without these data, there would be no scientific basis for establishing new standards or revising existing standards. Indeed, these networks were established in part to provide data for such research, as well as for assessing health and ecosystem impacts from air pollution, providing data for model evaluation, tracking pollutant trends, evaluating emerging pollutants, and assessing compliance with ambient standards and emission controls. Now, of course, determining effects of climate change has become another critical challenge requiring long-term measurements from robust and distributed networks. The importance of these networks in support of the EPA's mission of protecting and managing air quality is indisputable. Despite their value, these networks are faced perennially with uncertain funding and cutbacks that threaten the continuity of the critical data stream they provide. In May 2022, the EPA announced the termination or suspension of a number of sites associated with major national air quality monitoring programs, including CASTNET, National Trends Network (NTN), Ammonia Monitoring Network (AMoN) and Mercury Deposition Network (MDN) sites. Now in 2023, the EPA is proposing to close additional sites from CASTNET, which would leave major gaps in our understanding of air pollution under current and future air and climate policies. Rural monitoring sites in these networks provide air quality data in locations where state and local monitors rarely exist, making them an important resource for rural, low-income communities that often lack the funding and expertise to operate their own monitoring programs. Robust long-term air quality networks are critical to our understanding and protection of people, underserved communities and ecosystems during a period of changing climate. It is essential that they are supported and maintained to continue to support the protection of public health, natural resources, and sustainable economic growth. The CASAC recommends that EPA prioritize network funding for these and the other networks that come under its jurisdiction.

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Appendix A

Dissenting Opinion by Chartered CASAC Member Dr. James Boylan

One chartered CASAC member supports an alternative set of recommendations that are not based on the EAQM model results. The EAQM approach has not been peer reviewed and the results are highly questionable due to flaws in the conceptual model, simplifying assumptions, and missing processes leading to unreliable correlations between ambient air measurements and deposition values. Further, the results derived from the EAQM quantitative analysis generally show that correlations between local ambient measurements (SO₂, NO₂, and PM_{2.5}) and deposition values (S and N) are poor to nonexistent across all ecoregions, especially when looking at the more recent design values (2014-2016 and 2018-2020). As a result, it is not appropriate to use the EAQM modeling results in any form or fashion to set the secondary standards for SO₂, NO₂, and PM_{2.5}.

An alternative approach involves looking at historical TDEP values over the past 20 years. The PA documents the dramatic decrease in TDEP values after 2012. In fact, nearly all of the ecoregion 3-year average S and N deposition values within the last 10 years (2014-2016 and 2018-2020) are less than 5 kg/ha-yr and 10 kg/ha-yr, respectively. This happens to correspond with the promulgation and implementation of the 2010 1-hour SO₂, 2010 1-hour NO₂, and 2012 annual PM_{2.5} standards. This indicates that the current regulatory requirements are producing acceptable S and N deposition values in most parts of the county. It should be noted that there are still numerous areas that are not meeting these standards. As of August 31, 2023, 30 areas in the U.S. are nonattainment for the 2010 1-hour SO₂ standard, 5 areas are nonattainment for the 2012 annual PM_{2.5} standard, and 11 areas are nonattainment for the 2006 daily PM_{2.5} standard. No areas are currently nonattainment for the 2010 1-hour NO₂ standard. However, the current NO₂ monitoring network is not focused on large industrial NO_x point sources and violations of the 2010 1-hour NO₂ standard at some large industrial NO_x point sources have been demonstrated via dispersion modeling. Once all these areas are brought back into attainment with the current primary standards, there will be even greater reductions in S and N deposition values.

The current primary NAAQS for SO₂, NO₂, and annual PM_{2.5} (which are significantly more restrictive than the current secondary NAAQS for SO₂, NO₂, and annual PM_{2.5}) provide adequate protection against “long-term” annual S and N deposition-related effects. Therefore, the secondary NAAQS for SO₂, NO₂, and annual PM_{2.5} should be set equal to the current primary NAAQS for SO₂, NO₂, and annual PM_{2.5}.

Annual PM_{2.5} standard - Deposition-related Effects

Recommendation: 12 µg/m³ (annual average over three years)

Justification: Based on Figures 6-12, 6-13, 6-23, 6-24, 6-25, and 6-26, it appears that most of the S and N deposition values within the last 10 years are less than 5 kg/ha-yr and 10 kg/ha-yr, respectively. This indicates that the current primary annual PM_{2.5} standard adequately protects against “long-term” annual S and N deposition-related effects. The draft policy assessment discusses the option to set a secondary annual average PM_{2.5} standard as low as 12 µg/m³, which is the level of the current primary annual PM_{2.5} standard. Since the current primary PM_{2.5} NAAQS adequately protects against deposition related effects, the secondary PM_{2.5} NAAQS should be set equal to the current primary PM_{2.5} NAAQS.

1-hour NO₂ standard - Deposition-related Effects

Recommendation: 100 ppb (98th percentile of 1-hour daily maximum concentrations, averaged over 3 years).

Justification: Based on Figures 6-13, 6-20, and 6-21, it appears that most of the N deposition values within the last 10 years are less than 10 kg/ha-yr. This indicates that the current “short-term” primary 1-hour NO₂ standard adequately protects against “long-term” annual N deposition-related effects and can be used in lieu of setting a revised annual NO₂ standard. The draft policy assessment discusses the option to set a secondary annual average NO₂ standard as low as 40 ppb. However, the current “short-term” primary 1-hour NO₂ standard already provides protection for annual average NO₂ concentrations below 31 ppb (see Dr. Boylan’s individual comments). Since the current primary NO₂ NAAQS adequately protects against deposition related effects, the secondary NO₂ NAAQS should be set equal to the current primary NO₂ NAAQS.

1-hour SO₂ standard - Deposition-related Effects

Recommendation: 75 ppb (99th percentile of 1-hour daily maximum concentrations, averaged over 3 years).

Justification: Based on Figures 2-39, 6-12, 6-14, 6-15, 6-17, and 6-18, it appears that most of the S deposition values within the last 10 years are less than 5 kg/ha-yr. This indicates that the current “short-term” primary 1-hour SO₂ standard adequately protects against “long-term” annual S deposition-related effects and can be used in lieu of setting a new annual SO₂ standard. The draft PA discusses the option to set a secondary annual average SO₂ standard in the range of 10-22 ppb. However, the current “short-term” primary 1-hour SO₂ standard already provides protection for annual average SO₂ concentrations below 7.5 ppb (see Dr. Boylan’s individual comments). It should be noted that this recommendation is 25% lower than the lowest value proposed by the majority recommendation of 10-15 ppb annual average. Since the current primary SO₂ NAAQS adequately protects against deposition related effects, the secondary SO₂ NAAQS should be set equal to the current primary SO₂ NAAQS.

24-Hour PM_{2.5} standard - Deposition-related Effects

Recommendation: 35 µg/m³ (98th percentile of 24-hour concentrations averaged over 3 years)

Justification: Based on the fact that no analysis was presented in the PA relating the current 24-hour PM_{2.5} secondary standard of 35 µg/m³ or any alternative 24-hour PM_{2.5} secondary standards to S and N deposition, the current 24-hour standard should be retained without revision. It should be noted that this is consistent with the level proposed by the EPA in the most recent PM Policy Assessment to control for additional welfare effects.

Appendix B

**Individual Comments by the CASAC NO_x/SO_x/PM Panel on
EPA's *Policy Assessment for the Review of the Secondary National Ambient Air Quality
Standards for Oxides of Nitrogen, Oxides of Sulfur and Particulate Matter*
(External Review Draft – May 2023)**

Dr. Praveen Amar	B-2
Dr. James Boylan	B-6
Dr. Douglas Burns.....	B-13
Ms. Lauraine Chestnut.....	B-15
Dr. Judith C. Chow.....	B-17
Dr. Charles T. Driscoll, Jr.....	B-25
Dr. Ivan Fernandez.....	B-31
Dr. Frank Gilliam	B-37
Dr. Daven Henze	B-39
Dr. Robert W. Howarth.....	B-46
Dr. Donna Kenski.....	B-49
Dr. William McDowell.....	B-55
Dr. Hans Paerl.....	B-59
Mr. Richard L. Poirot.....	B-63
Dr. Alexandra Ponette-González.....	B-86
Dr. Stephen E. Schwartz	B-92
Dr. Kathleen Weathers	B-111

Dr. Praveen Amar

Chapter 1 - Introduction

To what extent does the Panel find the information in Chapter 1 is clearly presented and that it provides useful context for the review?

The introductory chapter provides a good background on the history of secondary standards starting from the year 1971 to the current 2023 PA. The following comment, however, is related to the need for technical/scientific editing of the complete PA document.

A main overarching comment on this and ALL the other chapters of PA is: This Policy Assessment document would benefit immensely if an external technical/science editor goes over the whole document. This Chapter notes on Page 1-3 (Lines 3 and 4) that this PA “is written to be understandable to a broad audience.” I do not believe that the current version of PA is “understandable to a broad audience”. It needs help of technical editors who have background and expertise in the field of effective “science communication”. For example, the PA has repetitive language in many chapters, such as many references to “uncertainty and limitations” that subtract from instead of add to the focus of this document. Another suggestion is to be parallel in use of words. For example, please say “oxides of nitrogen and oxides of sulfur” instead of nitrogen and sulfur oxides or N and SO_x or other similar words, when we should simply say “oxides of nitrogen and oxides of sulfur” every time. There are many other examples in this PA which do not use parallel language and would benefit by technical editing. Additionally, there is a general tendency in this PA to write many very long sentences that need to be broken down into clear and short sentences.

Page 1-1, Line 14: The “seven” chapters only list six chapters. Please include “Introduction” as one of the seven chapters.

Page 1-1, Line 22, please change the Title 1.1 to “The Purpose of Policy Assessment.”

Page 1-2, Line 11: We should clearly state the goal of the PA is to evaluate the public health and public welfare afforded by both the current and alternate future standards. Please say “..protection the current and alternate future standards afford.”

Page 1-2, Line 26: Please say “scientific information and risk and exposure assessment....”

Page 1-3: Lines 6-15: Please simply explain that “criteria pollutants” are named criteria pollutants simply because they are included in the “criteria documents.” Sounds obvious but not to the broad audience.

Page 1-4, Line 17: Please modify the Title to say “Oxides of Nitrogen and Oxides of Sulfur...” as noted earlier.

I think NAAQS was established for NO_x and NO₂ was simply the INDICATOR for NO_x. Is that correct? If yes, we should state it here.

Page 1-3 lines 29-30: Not clear what is meant by “EPA may consider “relative proximity to peak background...” A short explanation would be very helpful.

Page 1-6, Line15, please replace “CASAC felt” to more formal, “CASAC believed”.

Page 1-6, Line 35: I think we should simply say “scientific uncertainties: instead of “fundamental scientific uncertainties”. As noted earlier, we need to be careful in using the word “uncertainties” so many times. One suggestion is to write a separate section in this chapter (or, in Chapter 7 perhaps?) and discuss the subject of uncertainty in one place. The goal should not just list all kinds of uncertainties (in measurements, in models’ applications, in determination of various ecological effects, etc.) but how judgments must be made (and, are made) taking into account various uncertainties.

Page 1-8, Line 9: Please note the petitions were filed by several parties, RAISING a broad range of issues. The petitioners did not ADDRES the issues. They simply raised them.

Page 1-8, Line 30: not sure what is meant by the statement that EPA may consider “relative proximity to peak background concentrations”. I raised this issue above.

A general comment: It appears that more often than not, EPA has adopted the secondary NAAQS same as the primary NAAQS. It is not clear to me what is the the scientific and policy basis for these historic determinations.

Page 1-10, Lin3 16: What is meant by the word “generally” in the statement that EPA GENERALLY retained the 24-hr standard? Does it mean “not always”?

A general comment: It would be useful to note that in year 2012 when EPA decided not to adopt revised secondary standards because of uncertainties, it also proposed to undertake a five-year field program to collect atmospheric deposition and ecological data to lower the level of uncertainty in adopting any future secondary standards. What were the uncertainty-related findings of these field studies?

Page 1-13, Line 32: I do not think EPA produced REA (Risk Exposure Assessment) in 2020. I think EPA produced only a planning document (PD) for REA but REA itself was not undertaken. My understanding is that the current 2023 PA includes REA in it. Is that correct? If yes, please note this clearly in the current PA.

Page 1-14, Lines 9-14, and Lines 15-20: These two paragraphs are very unclear and confusing and need to be rewritten for clarity. Same for Lines 26-34. One suggestion is to break down long sentences into short sentences.

Page 1-15, Lines 1-7: Please include some more but brief information to make this paragraph more readable and meaningful. Many terms and concepts are mentioned here for the first time without clear explanations such as design value (DV), ecoregion, ecoregion air quality metrics (EAQM), air parcel trajectories, and zones of influence. All of these are important terms and are widely used in rest of the PA and need at least a brief paragraph or two here to provide a clear context.

Chapter 3 – Current Standards and Approach

To what extent does the Panel find that the background information in sections 3.1 and 3.2 to be clearly presented and provide useful context for the review? What are the Panel's views on the general approach described in section 3.3 for considering the welfare effects evidence and the risk assessment information for review of the adequacy of the current NO₂, SO₂, and PM secondary standards?

Chapter 3 provides a brief and useful context for this PA review.

My general comment is: How exactly this PA will address the “new” current deposition of sulfates, nitrates, and PM (and, if I may add, ammonia and ammonium (or, NH_x) deposition) in light of the historic baseline of large cumulative deposition of these pollutants to aquatic and terrestrial ecosystems? This issue is noted on Page 3-10 (Lines 11-14) and also at the end of this Chapter on Page 3-12 at Lines 18-26. I assume this issue might have been an important thing to consider during the 2012 Review (with discussion of AAI), but I do not recall if it was addressed then. My colleagues in this Panel are much more qualified to address this than me. This issue of course touches the issue of chemical (and, biological) recovery of the ecosystems so far under the previous emission controls (for example, Title IV of the 1990 Clean Air Act Amendments). For example, what more is needed for further recovery of these ecosystems under any alternate secondary standards? If this issue is discussed later in the PA, it should be noted here in this Chapter.

Another general comment /question I have is about many references in this Chapter and also in other chapters to the words “Quantitative Analyses”. (See Page 3-1, Lines 3-4). Please note that previous REA (Risk and Exposure Assessment) in the 2012 Review did undertake quantitative analyses of levels of risk to ecosystems at different exposure levels. My sense is that “Quantitative Analyses” has the same meaning as in “Risk and Exposure Assessment” (REA). Since this current review of the NAAQS did not perform a formal REA (a step that is between ISA and PA), do we mean REA when we say “Quantitative Analyses”? If they have the same meaning, we should state this clearly here or in Chapter 1. If not, we should state that too.

Page 3-5 Lines 4-6: it states ‘. that the general structure of an AAI-based standard addressed the potential for contributions to acid deposition from both oxides of nitrogen and of sulfur...’. However, in the 2012 review, the AAI-based standard also “took into account” reduced nitrogen deposition (NH₃, NH₄) in calculating AAI. Ammonia emissions were not targeted for control but the index stipulated how the role of ammonia would affect control of the other two pollutants (SOX and NO_x).

Page 3-6, Lines 1-3: One “overarching question” is noted here. However, later on Page 3-8, Line 33, it says that there are “overarching QUESTIONS” that are articulated in section 3.3” If there are more than one overarching question (many questions, perhaps), they should be listed here, so it is clear that PA is trying to address multiple overarching questions. For example, Chapter 7 lists a number of questions that need to be addressed before secondary NAAQS could be retained or revised. Are the questions listed in Chapter 7 the “remaining” overarching questions referred to in Section 3.3 but not listed in Chapter 3?

Page 3-10, Line 8: The end of the sentence “ ... such effects in air quality that meets the current standards” is awkward and should be rewritten for clarity. I know what you are trying to say but it does not say it clearly.

Page 3-11, Line 34: I think you mean “options” or “alternatives” instead of “considerations” here.

Page 3-12, Line 15: I think we should include here the effects of ammonia by saying "... the adequacy of protection from ecological effects afforded by the SO₂, NO_x, and PM secondary standards that also take into account the role of ammonia and ammonium deposition.

Dr. James Boylan

Chapter 2 – Air Quality

Some members of the panel have suggested that total light extinction may be a better indicator than total PM_{2.5} for S and N deposition. In theory this might be true, but I have not seen any comparisons of total light extinction vs. S and N deposition to confirm this.

Some members of the panel have suggested that this PA be expanded to cover visibility effects. However, I don't agree with asking EPA to evaluate visibility effects related to PM_{2.5} in this review since this review covers ecological effects of PM_{2.5} (visibility is not an ecological effect) and CASAC recently completed a detailed review of PM_{2.5} and visibility effects in early 2022. In 2023, EPA proposed the PM NAAQS rule that covers visibility impacts from PM_{2.5}. Any discussion of visibility effects are clearly outside the scope of this review.

Chapter 6 – Relationship of Deposition to Air Quality Metrics

There is a lack of peer reviewed published literature that looks at the relationship between ambient air concentrations and deposition with respect to the four elements of the NAAQS (indicator, averaging period, form, and level) for the current standard and alternative standards. As a result, EPA performed original work in the REA to inform the policy recommendations in the PA. The REA in Chapter 6 looks at the levels of protection provided by the current and alternative air quality standards. Since this REA work is not published or peer reviewed, the CASAC is responsible for providing feedback on that work and making recommendations as to the usefulness of the REA in standard setting. That is how the REAs have been done in all the CASAC reviews I have been involved with over the past 10 years (SO₂, ozone, PM, and this one). In my opinion, CASAC's review of the REA is a critical part of the overall NAAQS review process.

The scatter plots in Figures 6-4, 6-5, 6-9, 6-10, and 6-11 are very informative. However, there seems to be an issue with the units in some of the panels. In Figure 6-10, "S Deposition" ranges from 2-14 kg/ha-yr and "N Deposition" ranges from 1-15 kg/ha-yr, but "Total Deposition" ranges from 0-200 kg/ha-yr. In Figure 6-10, "IMPROVE N" ranges from 0-2.0 µg/m³, but "IMPROVE N+S" only ranges from 0-0.25 µg/m³. In Figure 6-11, "S Deposition" ranges from 0-30 kg/ha-yr and "N Deposition" ranges from 2-14 kg/ha-yr, but "Total Deposition" ranges from 0-300 kg/ha-yr. In Figure 6-11, "CASTNET TN" ranges from 0-2.0 µg/m³ and "CASTNET S" ranges from 0-5.0 µg/m³, but "CASTNET N+S" ranges from 0-300 µg/m³.

To better understand the relationship between the measured air quality concentrations and S and N deposition in various downwind locations of significance, EPA's EAQM assessment uses the HYSPLIT air parcel trajectory model to examine the transport of pollutant material from source to receptor. The HYSPLIT model is a peer-reviewed model that is commonly used for this type of analysis. It is stated on page 6-23 "The methodology used to calculate the air parcel trajectories that led to the sites of influence identification, as well as the methodologies used to estimate the EAQM values for each Ecoregion/pollutant pair using historical air quality design value (DV) data can be found in Appendix 6A." However, the methodologies used to estimate the EAQM values for each Ecoregion/pollutant pair are not clearly described in Appendix 6A.

The Appendix needs to include the equations that were used to calculate EAQM-weighted concentrations, EAQM-max concentrations, and the median S and N deposition values. The Appendix should explain why median deposition values were used instead of mean values. It seems that mean values might be more appropriate. Also, the Appendix should provide a more detailed explanation of how the number of “hits” were determined for each monitor? In addition, EPA should provide a simplified example showing how the equations are applied to generate the final EAQM values. In Figure 6A-1, it is strange to see grey circles in the middle of the Ecoregion since every trajectory originating from these monitoring locations will pass through large portions the Ecoregion and contribute to the deposition in that Ecoregion. In addition to a single figure showing a single ecoregion for a single pollutant (Figure 6A-1), the document should include maps of all the ecoregions and all the pollutants (SO₂, NO₂, and PM_{2.5}). Tables similar to Tables 6A-1 through 6A-4 need to be included for all combinations of ambient air pollutant concentrations and deposition values that were examined. In addition, these tables should include the number of SO₂, NO₂, and PM_{2.5} “monitoring sites of influence” that were included in the calculations for each Ecoregion.

Also, many of the assumptions need to be explained and justified. This includes:

1. EPA did not demonstrate that 1-year of meteorology is representative. EPA should run at least three years of meteorology to match number of years used to calculate design values. If not, EPA needs to create annual wind roses for 2016 and compare them to 3-year average wind roses.
2. It is not clear how many trajectories are released from each monitor location each day. Also, the start time for each trajectory is not included. This information should be added to the document.
3. It is not clear why a 500-meter trajectory height was used. Since the starting point for the trajectories are ambient monitoring sites located on the ground, it seems that 10-meter or 100-meter trajectories might be more appropriate.
4. It is not clear why EPA used 48-hour trajectories. Since sulfate can be transported long distances across multiple states, it might be more appropriate to use 72-hour or 96-hour trajectories.
5. It is not clear why EPA used a 1% contribution threshold. Additional justification should be provided. Also, EPA should consider 0.1% or 0.5% to increase the number of air quality monitors included in the analysis for each Ecoregion.

In addition to explaining and justifying the assumptions listed above, EPA should perform sensitivity runs using different assumptions to help quantify the uncertainty associated with this approach. Based on these sensitivity results, EPA should update the EAQM analysis and produce new results. **If the new EAQM results are significantly different than the results in the current PA, the CASAC should be given the opportunity to review a second draft of the PA.**

The conceptual model for this EAQM analysis consists of the following statements of fact and assumptions:

1. FACT – SO₂ and NO_x emissions are emitted from various sources (point, mobile, area, natural, etc.)
2. FACT – Ambient SO₂ and NO₂ monitors in each ecoregion are impacted by nearby SO₂ and NO_x emission sources.
3. ASSUMPTION – The current and historical SO₂ and NO₂ ambient monitoring network is adequate to represent the spatial gradients in each ecoregion.
4. ASSUMPTION – The majority of the S and N deposition in each ecoregion is from nearby SO₂ and NO_x emission sources.

5. FACT – SO₂ is converted to sulfate (SO₄ PM_{2.5}) and NO_x is converted to nitric acid (gas) and nitrate (NO₃ PM_{2.5}). Ammonia (gas) is converted to ammonium (NH₄ PM_{2.5}) in the form of ammonium sulfate, ammonium bisulfate, and ammonium nitrate.
6. FACT – Sulfate (SO₄ PM_{2.5}), nitrate (NO₃ PM_{2.5}), and ammonium (NH₄ PM_{2.5}) are part of total PM_{2.5}.
7. ASSUMPTION – Total PM_{2.5} measured at the surface is representative of aloft sulfate, nitrate, and ammonium associated with wet deposition.
8. QUESTION – Can nearby ambient measurements of SO₂, NO₂ and total PM_{2.5} be used to determine acceptable median S and N deposition values?

However, this model has several issues that need to be addressed:

1. The current ambient monitoring network for SO₂ does not capture the impacts from many of the large SO₂ industrial emission sources. Currently, there are two remaining coal-fired power plants in Georgia, but neither one has a nearby SO₂ monitor. Also, the current NO₂ ambient monitoring network was not designed to capture the impacts from large NO₂ industrial emission sources and is very scarce in many parts of the county (only three NO₂ monitors in Georgia, two near-road and one NCORE). In lieu of ambient monitoring, regulatory dispersion modeling has been performed for large industrial sources via the Data Requirements Rule (SO₂) and NSR permitting process (SO₂ and NO_x). As a result, the SO₂ and NO₂ ambient monitoring network may not be adequate to represent the spatial gradients in the Ecoregion.
2. EPA's EAQM approach does not account for atmospheric chemistry and the time it takes to convert SO₂ to sulfate and NO_x to nitrate and the fact that PM_{2.5} has long residence times and can be transported long distances. Recently, CAMx Particulate Matter Source Apportionment Technology (PSAT) modeling was performed for three Class I Areas in Georgia to support "Georgia's State Implementation Plan For Regional Haze (Second Planning Period), August 11, 2022" (<https://epd.georgia.gov/regional-haze-sip-second-implementation-period-0>). The PSAT modeling showed that only 5-10% of the sulfate and nitrate at Cohutta Wilderness Area (Figure 7-20, page 140), Okefenokee National Wilderness Area (Figure 7-21, page 140), and Wolf Island National Wilderness Area (Figure 7-22, page 141) came from sources in Georgia while the remaining 90-95% came from sources outside Georgia. For Cohutta Wilderness Area (Table 7-16, page 181), the PSAT results for the eight individual facilities with sulfate contributions greater than 1% are shown below.

State	Facility ID	Facility Name	DISTANCE_km	FINAL Revised Sulfate PSAT Mm ⁻¹	FINAL Revised EGU + NEGU Mm ⁻¹	FINAL Revised Sulfate PSAT %
OH	39053-8148511	General James M. Gavin Power Plant (0627010056)	512.0	0.322	13.229	2.44%
GA	13015-2813011	Ga Power Company - Plant Bowen	78.0	0.282	13.229	2.13%
KY	21145-6037011	Tennessee Valley Authority (TVA) - Shawnee Fossil Plant	457.2	0.190	13.229	1.44%
IN	18147-8017211	INDIANA MICHIGAN POWER DBA AEP ROCKPORT	410.1	0.181	13.229	1.37%
OH	39025-8294311	Duke Energy Ohio, Wm. H. Zimmer Station (1413090154)	454.6	0.173	13.229	1.31%
TN	47163-3982311	EASTMAN CHEMICAL COMPANY	269.8	0.165	13.229	1.25%
PA	42005-3866111	GENON NE MGMT CO/KEYSTONE STA	801.1	0.137	13.229	1.04%
IN	18051-7363111	Gibson	487.1	0.137	13.229	1.03%

It should be noted that the only source in Georgia with a contribution greater than 1% is Georgia Power-Plant Bowen, which does not have a nearby SO₂ ambient monitor. The other seven facilities with sulfate contributions greater than 1% are located in six different states and are 270 – 801 km away from the Cohutta Wilderness Area. If the majority of the sulfate and nitrate is being transported from far outside the ecoregion, then the use of nearby SO₂ and NO₂ monitors inside the ecoregion to evaluate S and N deposition does not seem appropriate.

- EPA's EAQM approach does not account for the significant contributions of nitric acid wet and dry deposition to N deposition.
- Total PM_{2.5} is a poor indicator of sulfate (SO₄ PM_{2.5}), nitrate (NO₃ PM_{2.5}), and ammonium (NH₄ PM_{2.5}). Twenty years ago, total PM_{2.5} might have been an acceptable indicator when the majority (60-75%) of total PM_{2.5} was sulfate and nitrate. However, based on the 2019-2021 speciated PM_{2.5} data in Figure 2-26, only 20-40% of total PM_{2.5} is sulfate and nitrate. If you convert the OC to OM (by multiplying OC*1.4) and include the unidentified PM_{2.5} mass (typically OM), then the percent contributions from sulfate and nitrate drop to 15-30%. Also, these percentages are even smaller on days that exceed the current 24-hour standard since the additional PM_{2.5} mass on those days are typically organic carbon and elemental carbon from wildfires and prescribed fires, or crustal material from dust events. Total PM_{2.5} measured at the surface is not representative of sulfate, nitrate, and ammonium at the surface; therefore, it would likely not be representative of aloft sulfate, nitrate, and ammonium associated with wet deposition.

In my opinion, the EAQM analyses for SO₂ and NO₂ are highly questionable due to flaws in the conceptual model, simplifying assumptions, and missing processes leading to unreliable correlations between ambient air measurements and deposition values. Also, the EAQM analyses for PM_{2.5} are questionable due to the fact that total PM_{2.5} is not a good indicator for S and N deposition and nitric acid is not accounted for in the analysis. As a result, I do not believe it is appropriate to use the EAQM modeling results to set the secondary standards for SO₂, NO₂, and PM_{2.5}. However, I think the EAQM

analysis might be appropriate if applied to: (1) sulfate ($\text{SO}_4 \text{ PM}_{2.5}$) vs. S deposition, (2) nitrate ($\text{NO}_3 \text{ PM}_{2.5}$) + ammonium ($\text{NH}_4 \text{ PM}_{2.5}$) vs. N deposition, or (3) nitrate ($\text{NO}_3 \text{ PM}_{2.5}$) + ammonium ($\text{NH}_4 \text{ PM}_{2.5}$) + nitric acid (HNO_3 gas) vs. N deposition. These analyses should be considered during the next NAAQS review.

Also, the generally poor correlations between the ambient measurements (SO_2 , NO_2 , and $\text{PM}_{2.5}$) and deposition values (S and N) provides yet another reason that EAQM modeling results should not be used to set the secondary standards for SO_2 , NO_2 , and $\text{PM}_{2.5}$. This is especially true when looking at the more recent design values (2014-2016 and 2018-2020) compared to older design values (2001-2003, 2006-2008, and 2010-2012) and the Western U.S. compared to the Eastern U.S.

Chapter 7 – Review of the Standards

Most of the secondary NAAQS recommendations in this Chapter are based on the EAQM work presented in Chapter 6. Since that modeling work has not been thoroughly peer reviewed, the results should be used with extreme caution. Based on the numerous EAQM shortcomings that were identified in Chapter 6 and the poor correlations between ambient measurements of SO_2 , NO_2 , and $\text{PM}_{2.5}$ and S and N deposition values, I do not believe it is appropriate to use the EAQM modeling results that are presented in the draft policy assessment to set the secondary standards for SO_2 , NO_2 , and $\text{PM}_{2.5}$.

3-hour SO_2 standard - Direct Effects

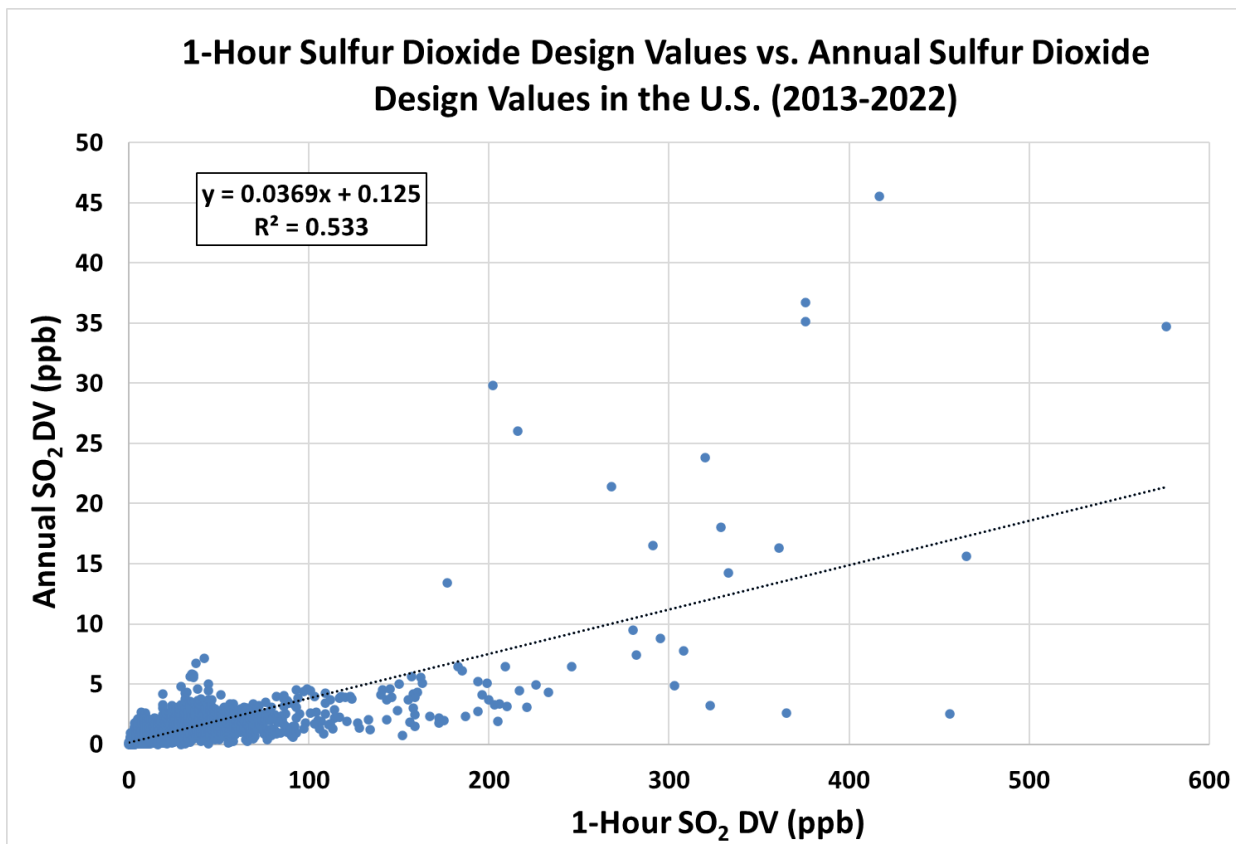
***Recommendation:* 0.5 ppm (3-hour maximum not to be exceeded more than once per year)**

Justification: The current standard of 0.5 ppm is adequate to protect against direct effects and should be retained.

1-hour SO_2 standard - Deposition-related Effects

***Recommendation:* 75 ppb (99th percentile of 1-hour daily maximum concentrations, averaged over 3 years)**

Justification: Based on Figures 2-39, 6-12, 6-14, 6-15, 6-17, and 6-18, it appears that most of the S deposition values within the last 10 years are less than 5 kg/ha/yr. This indicates that the current “short-term” primary 1-hour SO_2 standard adequately protects against “long-term” annual S deposition-related effects and can be used in lieu of setting a new annual SO_2 standard. The figure below compares 1-hour SO_2 design values against annual SO_2 design values for all valid SO_2 design value pairs in the U.S. from 2013-2022.



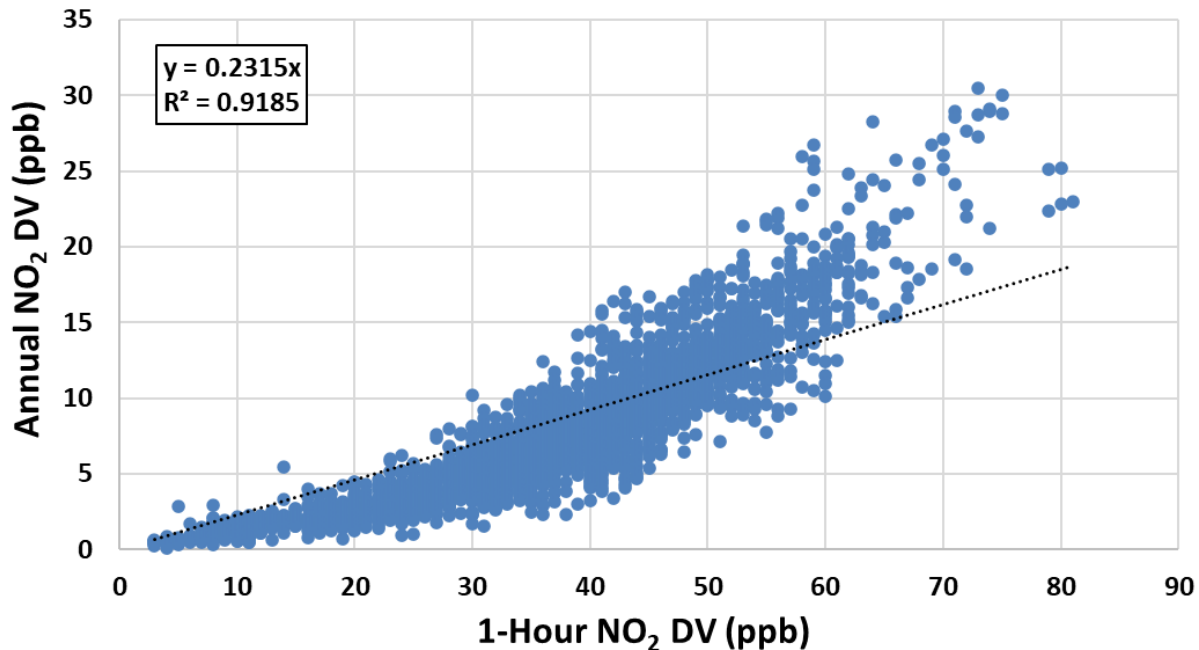
The draft policy assessment discusses the option to set an annual average SO₂ standard in the range of 10-22 ppb. However, the current “short-term” primary 1-hour SO₂ standard already provides protection for annual average SO₂ concentrations below 7.5 ppb. Therefore, the secondary SO₂ NAAQS should be set equal to the current primary SO₂ NAAQS to protect against deposition related effects.

1-hour NO₂ standard - Deposition-related Effects

***Recommendation:* 100 ppb (98th percentile of 1-hour daily maximum concentrations, averaged over 3 years)**

Justification: Based on Figures 6-13, 6-20, and 6-21, it appears that most of the N deposition values within the last 10 years are less than 10 kg/ha/yr. This indicates that the current “short-term” primary 1-hour NO₂ standard adequately protects against “long-term” annual N deposition-related effects and can be used in lieu of setting a revised annual NO₂ standard. The figure below compares 1-hour NO₂ design values against annual NO₂ design values for all valid NO₂ design value pairs in the U.S. from 2013-2022.

1-Hour Nitrogen Dioxide Design Values vs. Annual Nitrogen Dioxide Design Value in the U.S., 2013-2022



The draft policy assessment discusses the option to set an annual average NO₂ standard as low as 40 ppb. However, the current “short-term” primary 1-hour NO₂ standard already provides protection for annual average NO₂ concentrations below 31 ppb. Therefore, the secondary NO₂ NAAQS should be set equal to the current primary NO₂ NAAQS to protect against deposition related effects.

Annual PM_{2.5} standard - Deposition-related Effects

***Recommendation:* 12 µg/m³ (annual average over three years)**

Justification: Based on Figures 6-12, 6-13, 6-23, 6-24, 6-25, and 6-26, it appears that most of the S and N deposition values within the last 10 years are less than 5 kg/ha/yr and 10 kg/ha/yr, respectively. This indicates that the current primary annual PM_{2.5} standard adequately protects against “long-term” annual S and N deposition-related effects. Therefore, the secondary PM_{2.5} NAAQS should be set equal to the current primary PM_{2.5} NAAQS to protect against deposition related effects.

24-Hour PM_{2.5} standard - Deposition-related Effects

***Recommendation:* 35 µg/m³ (98th percentile of 24-hour concentrations averaged over 3 years)**

Justification: Based on the fact that no analysis was presented in the PA relating the current 24-hour PM_{2.5} secondary standard of 35 µg/m³ or any alternative 24-hour PM_{2.5} secondary standards to S and N deposition, the current 24-hour standard should be retained without revision. It should be noted that this is consistent with the level proposed by EPA in the most recent PM Policy Assessment to control for additional welfare effects.

Dr. Douglas Burns

Chapter 5 – Exposure Conditions Associated with Effects

Does the Panel find the presentation of information and quantitative analyses related to welfare effects of oxides of nitrogen, oxides of sulfur and particulate matter to be technically sound and clearly communicated? What are the Panel's views on the quantitative analysis of aquatic acidification risks, including the choice of analysis approach and spatial resolutions? Does the document appropriately characterize the results of the analysis, including key limitations and associated uncertainties?

I found the information and analyses related to ecosystem effects of NO_x, SO_x, and particulates to be generally technically sound and understandably communicated. Some of the deposition CL exceedance pairs analysis gets a little dense and tedious to read but does present the available CL analyses in a comprehensive manner. Because this review has stretched over many years, new data and studies after about 2017/18 are not presented. While there have not been large changes in scientific understanding over this period, there have been many new studies on CLs, some using modern machine learning approaches that have reduced uncertainties. Given the completion date of the ISA in about 2018, there is probably nothing to be done about this shortcoming, but I suspect that some in the CASAC panel will want full consideration to be given to new observations and modeling results.

The only areas of technical concern as I read this chapter was the extended discussion of soil BC/Al ratios as a threshold indicator of effects on vegetation. I believe that this indicator has been largely proven to be of value only in a qualitative manner as there are too many exceptions that limit its value as a quantitative indicator. One final comment is that I was expecting some discussion of the role of particulate forms of nitrogen as these contribute to overall total deposition. Especially in arid regions, particulate N is a very large contributor to the general ecosystem effects of atmospheric deposition.

Regarding the quantitative analysis of aquatic acidification, I found this analysis to be generally sound and useful in terms of spatial resolution. The document does a thorough job of describing the results, limitations, and uncertainties. ANC is the most useful metric to apply despite the influence of other factors such as natural organic acidity on values. The authors dealt with this by eliminating from further consideration, areas such as the coastal plain where organic acidity is dominant. I understand the justification for not including N deposition in the CL exceedance analysis because of the necessary averaging of ANC values at sites over an extended time period. However, streams and lakes in the northeastern US generally show episodic acidification in the winter and early spring during snowmelt and this phenomenon is most often driven by nitrate. Because this analysis does not really consider episodic acidification in a CL exceedance framework (despite discussion in the text of the role of episodic acidification in harming fish and other aquatic organisms), nitrate and N deposition appear to be less important than in fact they really are to episodic ecosystem effects.

Below are some bulleted comments linked to specific locations in the text. Many of these are minor concerns but I wanted to express where I felt the text had some mostly minor shortcomings or requires editorial corrections.

- Page 5-6, L33 – worth stating that there are several landscape factors that are important here such a slope steepness, drainage density, and extent of riparian area among others
- Page 5-10, L5 – should say insensitive here

- Page 5-13 – regarding the conceptual model, NH₃ could be added as an atmospheric deposition component, reduced biodiversity important as well as biomass
- Page 5-19, L24 – sulfur is a nutrient as well and subject to uptake just as nitrogen. Perhaps the nutrient role of sulfur can be set aside because the supply has historically been so high relative to biological demand, but that is changing in recent years
- Page 5-53, L12 – C/N ratios have been identified as a diagnostic soil indicator of tendency for nitrate leaching---see the work of Christine Goodale and others
- Page 5-54 – I was surprised to see such an extensive discussion of soil BC/Al as most soil biogeochemists working in this field would today conclude that it is not a very strong diagnostic indicator and it is particularly difficult to establish clear threshold values as in aquatic CL studies. Scientists such as Greg Lawrence, Scott Bailey, and others would suggest that other metrics such as percent base saturation and exchangeable calcium are better indicators.
- Page 5-56 – no mention here of methods using aquatic chemistry to estimate cation weathering rates. Shaun Watmough and Scott Bailey are two scientists who have published studies on these approaches to estimate base cation weathering rates.
- Page 5-60, L3 – text should say “at/below”

Chapter 6 – Relationships of Deposition to Air Quality Metrics

To what extent does the Panel find the investigation of relationships between air concentration and deposition to be technically sound and clearly communicated? What are the CASAC views on the consideration of key limitations and associated uncertainties?

The analyses in chapter 6 show somewhat strong relationships between air concentrations and deposition, but also reflect considerable uncertainty when attempting for example to link a CL value directly to air concentrations of SO₂ and NO₂. First, the lack of measurements of reduced N in current networks results in weaker relationships for N than S deposition. Bringing reduced N into the estimates via CMAQ simulations considerably improves the relationship. Another concern is that the air concentration – deposition relationship appears to be weakening in recent years as deposition levels decline. This adds additional uncertainty to establishing precise secondary air quality standards. I agree with the authors that using a 3-year annual average makes more sense than a maximum value, especially with consideration that long-term deposition levels are largely what determine ecosystem effects. Perhaps air concentration values that are never exceeded in the analysis relative to a CL threshold would be the best approach as seems to be implied in the chapter text.

Ms. Lauraine Chestnut

Chapter 1 – Introduction

Overall, this chapter provides a good and succinct accounting of the previous reviews of the secondary NAAQS for SO_x, NO_x, and PM and sets an appropriate context for this document. There are, however, a few places where a bit more information would be helpful.

Pages 1-4 to 1-6: What specifically are the vegetation effects that the current standards protect against? This can be brief and descriptive. Do the standards prevent death of vegetation, reduced growth and productivity? Are there certain species or certain locations that are the primary recipients of the protection?

Page 1-7: What was the rationale for setting the PM_{2.5} secondary standard equal to the primary? The next page mentions visibility; was that the main consideration?

As further context for this review, I think something should be said about the Acid Rain Program under the 1990 Clean Air Act Amendments. I realize that this is a different regulatory process than the NAAQS but because it was specifically motivated by wanting to reduce the recognized harm being done by acid deposition and because it has had such a significant effect on reducing precursor emissions, especially SO₂, it seems like it should be discussed in this chapter that sets the context for this review. That Congress included this in the amendments is strong support that adverse public welfare effects of acid deposition were recognized at that time.

Chapter 3 – Current Standards and Approach

This chapter provides a good overview of the current secondary standards for NO_x, SO_x, and PM, and the previous review of potential secondary standards for NO_x and SO_x as they affect ecosystems.

Page 3-2: The paragraph about the PM secondary standards could use a bit more specificity. The last sentence implies that the review when these secondary standards were set did take into consideration the ecological effects of PM and that these standards protect against these effects. Is that accurate?

Chapter 4 –Nature of Welfare Effects

Section 4.3 provides a very good general discussion of public welfare effects. The section rightly notes that it is up to the Administrator to decide if the effects rise to the level of adverse, but more could be said to help in this decision. The last row in Figure 4.3 can be much more assertive about describing what are the public welfare effects of deposition. These are not “possible”; there is a lot of confidence that these effects happen, and they fall within the definition of public welfare effects. These effects span direct human usage and enjoyment of the environment and the fact that people care about protection of biodiversity and healthy habitats even if they never visit these places. Bringing forward some of the discussion of ecosystem services from the ISA may be helpful here. What is needed is to make clear why the effects described in the previous sections are important. More specific summary descriptions would help emphasize the results discussed in the previous sections. For example, say that acidification of

streams and lakes (in ___ regions) has resulted in loss of sensitive species of fish and other biota, including fish species popular with anglers. Further, effects don't have to be fully quantified for us to be confident that they are happening. For example, it may not be possible to say the specific amount that N deposition is contributing to eutrophication in an estuary, but we may be quite confident that it is contributing and therefore causing associated public welfare effects.

Chapter 7 – Review of the Standards

This chapter correctly notes that it is up to the Administrator to make the judgement about what standards may be requisite to protect against adverse welfare effects, but I think more could be brought forward from the previous chapters to support the Administrator in making this judgement. For example, brief descriptive information about the nature and extent of the effects that each standard would protect against would be helpful.

There is so much emphasis on the uncertainties that it is hard sometimes to keep in mind what is reasonably well known and understood.

Additional discussion would help in assessing the different amounts of protection that would be afforded by the different levels of standards being proposed for consideration. Are there any differences in the effects that would be protected against over the range of standards being considered? How would you characterize the risk of over or under protecting over the range of standards being considered?

I think it would also be helpful to say how each of these proposals compares to current concentrations. Are proposed standards these already met in most locations?

Dr. Judith C. Chow

Chapter 2 –Air Quality

To what extent does the Panel find that the information in Chapter 2 is clearly presented and that it provides useful context for the review?

Chapter 2 is well written and documents atmospheric processes, emissions, measurement methods, recent trends, and deposition of nitrogen (N), sulfur (S), and particulate matter (PM). Following are a few suggestions for clarification.

2.1 Atmospheric Transformation of Nitrogen, Sulfur, and PM species

This section briefly describes the atmospheric processes (Section 2.1.4) without acknowledging the non-linear relationship between the precursor gases and secondary aerosol formation (e.g., Blanchard and Hidy, 2003; Blanchard and Hidy, 2005; Blanchard et al., 2007; Stockwell et al., 2000). Pathways for reactive nitrogen species are shown in Figure 2-5 of the ISA under Section 2.3 of Atmospheric Chemistry of Nitrogen and Sulfur Species and Particulate Matter (U.S.EPA, 2020, page 2-18), but it lacks explanation of associations with particulate nitrate (NO_3^-).

Nitrate chemistry is more complicated than, and dependent on, sulfate (SO_4^{2-}) chemistry. In addition to converting NO_x emissions to nitric acid (HNO_3) and peroxyacetyl nitrate (PAN), there are other reactive species (e.g., OH, O_3 , HNO_3 , and N_2O_3) that influence atmospheric processes. The major pathway for HNO_3 formation involves the same hydroxyl radicals that transform sulfur dioxide (SO_2) to sulfuric acid (H_2SO_4). Unlike ammonium sulfate, ammonium nitrate is not stable in the atmosphere. Its equilibrium with gaseous ammonia (NH_3) and HNO_3 is temperature and relative humidity dependent. As shown in the Figure A example below, the extent of ammonium nitrate reduction is a function of the total nitrate ($\text{HNO}_3 + \text{NO}_3^-$) and total ammonia ($\text{NH}_3 + \text{NH}_4^+$) (Watson et al., 1994).

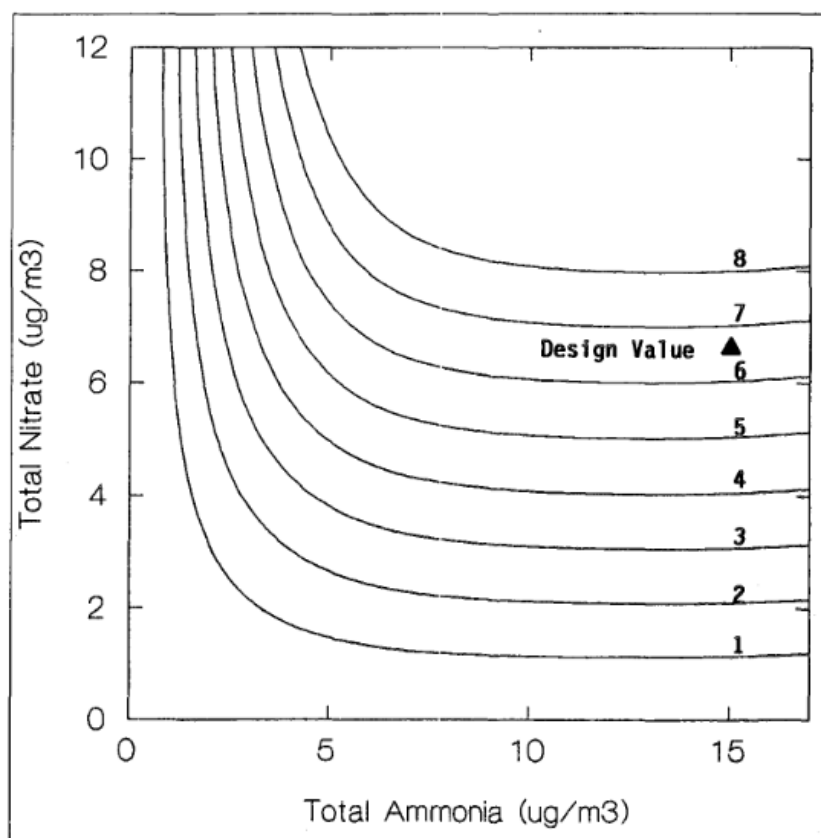


Figure A. Particulate nitrate isopleths for low relative humidity conditions on the morning of December 26, 1989 in Phoenix, AZ. The number on each isopleth is the ammonium nitrate concentration in $\mu\text{g}/\text{m}^3$. The design value corresponds to the total nitrate and total ammonia equivalents (gas plus particle) measured for this sample (Watson et al., 1994). This shows that ammonia reductions have negligible effect on ammonium nitrate formation until they are reduced by $\sim 70\%$.

These policy-relevant concepts need to be stated, as effective particulate emission control strategies may vary by geographical regions and different meteorological conditions.

2.3 Monitoring Ambient Air Concentrations and Deposition of N, S, and PM

Section 2.3 documents NO_x , SO_2 , and $\text{PM}_{2.5}$ monitoring networks (Figures 2-11, 2-12, and 2-13 from Pages 2-15 to 2-17 respectively). It would be informative to include a map with collocated NO_x , SO_2 , and $\text{PM}_{2.5}$ sites. Note that sampling frequencies (Line 12, page 2-17) for the Chemical Speciation Network (CSN) and Interagency Monitoring of Protected Visual Environments (IMPROVE) sites are incorrect. IMPROVE sites sample every third day on the U.S.EPA (2023) schedule while many (but not all) of the CSN sites sample every sixth day, with the exception of NCore sites that sample every third day.

Section 2.3.4 “Other Monitoring Networks Relevant to N, S, and PM Deposition” shows locations in the 250 National Acid Deposition Program/National Trends Network (NADP/NTN) sites. However, Figure 2-15 (Page 2-19) is illegible, a smaller symbol size for each site would help to discern their general locations. If the Ammonia Monitoring Network (AMoN) (Figure 2-12, Page 2-22) is part of NADP, locations of collocated AMoN and NADP sites should be identified.

Section 2.3.4 notes that speciated measurements in the Clean Air Status and Trends Network (CASTNET) include O_3 , SO_2 , HNO_3 , NO_3^- , SO_4^{2-} , and NH_4^+ among others (Lines 1 and 2, Page 2-21). Other anion (i.e., Cl^-) and cations (Mg^{++} , Ca^{++} , and Na^+) acquired by CASTNET should be noted. In addition, hourly nitric oxide (NO), reactive nitrogen (NO_y), and CO are also acquired at the CASTNET sites (Feng et al., 2020), as well as at NCore locations. These measurements can be used to estimate acid neutralizing capacities (ANC) for evaluation of aquatic biota effects and as basis for calculating critical loads (CL) for forest soils and surface waters.

The statement that “Due to large measurement artifacts, IMPROVE no longer reports ammonium (NH_4^+)...” (Lines 24 and 25, Page 2-21) is incorrect. Cations were never part of the IMPROVE measurements except during special studies. The IMPROVE reconstructed mass formula (Chow et al., 2015) assumes that all nitrate is in the form of ammonium nitrate, which appears to be valid in all but coastal areas where sodium nitrate may be a contributor.

The 24-hour standard for $\text{PM}_{2.5}$ is $35 \mu\text{g}/\text{m}^3$, not $150 \mu\text{g}/\text{m}^3$ (Lines 4 and 5, Page 2-29). The pie chart map shown in Figure 2-26 (Page 2-29) includes SO_4^{2-} , NO_3^- , OC, EC, sea salt, and crustal components, but this does not represent reconstructed $\text{PM}_{2.5}$ mass. Measured or calculated NH_4^+ (based the assumption that ammonium nitrate and ammonium sulfate or ammonium bisulfate are present when ammonium is not measured) (Chow et al., 1994) should be included as separate components. Since organic material (OM) does not account for unmeasured oxygen, hydrogen, sulfur, and nitrogen, factors of 1.2 to 1.8 that are used to convert OC to OM, and these ratios appear to be changing over time (Hand et al., 2019). The difference between $\text{PM}_{2.5}$ mass and reconstructed mass should be included as “unidentified material” (possibly liquid water, artifact, uncertain multipliers, or unknown contributors) to indicate potential uncertainties.

2.4 Recent Ambient Air Concentrations and Trends

Average NO_3^- and SO_4^{2-} concentrations in the US for the period of 2019-2021 are presented in Figures 2-29 and 2-30 (Page 2-32) with annual trends (2006 to 2021) shown in Figures 2-31 and 2-32 (Page 2-33). The sources of these data should be identified in the captions. Similar figures for spatial distributions and long-term trends for measured NH_4^+ levels at CSN and CASTNET sites and estimated NH_4^+ from the NO_3^- and SO_4^{2-} levels at IMPROVE sites would be useful.

Section 2.4.3 “ $\text{PM}_{2.5}$ Concentrations and Trends” notes that “Nationally, annual average $\text{PM}_{2.5}$ concentrations have declined despite the relatively consistent trends in NH_3 emissions” (Lines 17-18, Page 2-30). This statement contradicts the upward trends in NH_3 emissions from ~4000 to 5000 k tons/yr from ~2017 to 2020 shown in Figure 2-10 (Page 2-12). The “other anthropogenic sources” need to be specified in this Figure. However, Section 2.5.4 “Trends in Deposition” indicates that changes in total nitrogen deposition are a combination of declining oxidized nitrogen and increasing reduced nitrogen (Lines 13-14, Page 2-16); some consistency across sections is needed.

Butler et al. (2016) shows increasing air NH_3 and precipitation NH_4^+ over larger regions in the U.S. from 2008 to 2015. Reduced SO_2 and NO_x emissions resulted in less H_2SO_4 and HNO_3 for NH_3 neutralization in the atmosphere. This type of data analysis is needed to better understand the spatial variations of NH_3 and NH_4^+ under different meteorological conditions in different U.S. regions.

In addition to deposition, visibility should be considered as part of welfare effects in secondary PM, as noted in the Code of Federal Regulations (CFR, 2023). The IMPROVE network was initiated in 1987/88

to identify and evaluate visibility impairment or deterioration in Class I areas. As ammonium sulfate and ammonium nitrate are major light scattering components in PM_{2.5}, it would be informative to include trends on visibility or chemical light extinction for the 20% most impaired days in recent years.

2.5 Nitrogen and Sulfur Depositions

The number and locations of collocated CASTNET and AMoN sites should be documented. For reduced nitrogen components, were weekly averaged CASTNET NH₄⁺ concentrations combined with collocated two-week averaged AMoN NH₃ concentrations for total deposition (TDEP) model evaluations? Figure 2-40 (Page 2-45) shows spatial variations (in average percentages) of reduced nitrogen deposition. For the period of 2019-2021, percent distributions for reduced nitrogen (Figure 2-40) generally followed the pattern of total nitrogen deposition (KgN/ha) as shown in Figure 2-38 (Page 2-43). Similar graphs for total, wet, and dry nitrogen deposition should be included for comparison.

Table 2-26 (Page 2-47) summarizes changes in total deposition between the 2000-2002 and 2019-2021 periods for total oxidized and reduced nitrogen deposition by region (a footnote is needed to show states/areas for each of the eight regions). For the North Central region, total deposition of oxidized nitrogen and reduced nitrogen increased by 56% and 11%, respectively, resulting in a 37% reduction in total deposition nitrogen. The largest increase in reduced nitrogen (+111%) is found in South Central region (consistent with the increases in annual emission trends), along with 39% reduction for total oxidized nitrogen, resulting in a 16% increase in total nitrogen deposition. These examples demonstrate the non-linear nature of nitrate chemistry.

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Chapter 7 – Review of the Standards

What are the Panel views on the discussion of key considerations in sections 7.1 and 7.2 preliminary conclusions?

Chapter 7 adequately summarizes available information relevant to potential welfare effects. It is apparent that longer averaging times would better represent deposition-related effects on ecosystems. However, relationships between the proposed concentration range and ecosystem effects need better documentation.

A rationale for the 200-400 ppb range for the second highest daily 3-hr maximum SO₂ with respect to deposition-related effects needs to be provided. Figure 2-23 (Page 2-27) shows that all sites have SO₂ secondary standard design values <500 ppb, with 78% of the sites (274 out of 351) having concentrations <20 ppb, and 1.1% (4 sites) with design values between 201-500 ppb. Section 7.3 “Preliminary Conclusion” states that 95% of the maximum 3-hr average concentrations have been less than 200 ppb and 99% are below 300 ppb since 2000 (with the exception of Hawaii, with high values attributable to volcanic activity) (Lines 5-8, Page 7-25). A proposed range of 200-300 ppb would be justifiable for the 3-hr average secondary SO₂ NAAQS.

The three year annual SO₂ concentrations of 10-22 ppb (Table 7-1, Page 7-32) have been associated with median S deposition rates of ~5 kg/ha-yr in the Ecoregion III areas (Lines 3-5, Page 7-15). Section 6.2.5 (Conclusions) shows that the majority of the monitors registered below 10 ppb for the period of 2018-2020 (Lines 14-17, Page 6-36). The proposed SO₂ range is much higher than the annual average SO₂ design values shown in Figure 2-25 (Page 2-28) with <5 ppb since 2001 and <1 ppb for 2019-2021. A level of 10 ppb would be more appropriate for the 3 year annual average secondary SO₂ standard.

The rationale for reducing the annual NO₂ secondary NAAQS limit from 53 ppb to 40 ppb also needs elaboration. Section 2.4.1 “NO₂ Concentrations and Trends” shows that average hourly NO₂ values were 16.3 ppb for all sites (Lines 9-13, Page 2-23). There has been no exceedance of the NO₂ NAAQS since 1991. Annual mean NO₂ design values shown in Figure 2-21 (Page 2-25) range from ~23 ppb in 1980 to ~8 ppb in 2021. Primary and secondary NO₂ single year design values show that over 98% of the sites (396 out of 404 sites) measure below 20 ppb (Figure 2-19 Page 2-24). Figure 6-21 (Page 6-31) shows that recent (2018-2020) median N deposition rates of <12 kg/ha-yr in Ecoregion III areas are associated with 3-year annual average NO₂ design values <30 ppb. The scatterplot of 3-year average N deposition vs. weighted secondary NO₂ design values in Figure 6-20 (Page 6-30) shows <15 ppb annual average NO₂ since 2014. The proposed 40 ppb would not effectively alter deposition-related effects. A level of ≤20 ppb seems more appropriate for the annual secondary NO₂ NAAQS.

Table 7-1 would be more informative if the proposed secondary NO_x/SO_x/PM NAAQS levels that can be associated with median S and N depositions (kg/ha-yr). Approaches for protecting public welfare are associated with acid neutralizing capacity (ANC) and critical loads (CL). The relationship between the proposed secondary SO₂ and NO₂ NAAQS levels and waterbody ANC levels as well as relevant deposition amounts (kg/ha-yr) per ecoregion need elaboration to justify the protection of aquatic biota from acid deposition.

The proposed 3-year average annual PM_{2.5} standard also needs to be associated with S and N deposition rates. Figures 6-12 and 6-13 (Page 6-21) show that S and N depositions are associated with <10 µg/m³ PM_{2.5} mass for the period of 2014-2019. Estimated 3-year average “S+N deposition” values in Figure 6-28 (Page 6-35) are also associated with a weighted average of PM_{2.5} of 10 µg/m³ for the period of 2014-2016. For the period of 2018-2020 there are a few points scattered between 10 to 12 µg/m³. However, the concentrations in this range are associated with low deposition rates (Figure 6-28). The secondary PM_{2.5} standard of 10 µg/m³ is also consistent with the recent CASAC PM Panel findings that recommend an annual PM_{2.5} NAAQS in the range of 8-10 µg/m³.

As the annual PM_{2.5} NAAQS does not necessarily protect against episodic events (e.g., wintertime stagnation, residential wood combustion, and summer/fall wildfires), EPA is encouraged to change the 24-hr secondary PM_{2.5} standard from 35 to 25 µg/m³. This is in line with a visibility index of 25 deciviews, a level that is accepted by ~50% of the preference study participants. The 25 µg/m³ also falls into the range of 25-30 µg/m³ recommended by the CASAC PM Panel for primary 24-hour PM_{2.5} NAAQS. More data analysis is needed for areas with 24-hr PM_{2.5} concentrations exceeding 25 µg/m³ to explore their relationship with S and N depositions and visibility impairment by SO₄²⁻, NO₃⁻, and NH₄⁺.

What are the CASAC views regarding the areas for additional research identified in Chapters 3 and 4? Are there additional areas that should be highlighted?

Section 7.4 “Areas for Future Research Related to Key Uncertainties” provides a good list of data acquisition and model development efforts to address the limitations and uncertainties of current scientific evidence. One key factor to improve model estimates is the need for adequate model inputs and a corresponding measurement network for model evaluations. Networks were established at various times and are managed/operated/funded by different federal/state agencies, with many (but not all) non-collocated measurement locations, different sample durations and frequencies, and different data formats. A multipollutant air quality management approach (e.g., Chow et al., 2010; Chow and Watson, 2011; Hidy et al., 2017) needs to be considered to address multipollutant effects.

Sampling locations (e.g., urban, suburban, and rural); sampling methods (e.g. continuous gas vs. integrated filter pack sampling, active CASTNET sampling vs. passive [Radiello sampler] AMoN); sampling durations (e.g., 24 hr at CSN and IMPROVE vs. one week for CASTNET and two weeks for AMoN); and sampling frequencies (e.g., continuous hourly for NO₂, SO₂, and O₃, every third day for IMPROVE, and every third or sixth day for CSN) need to be synchronized to the extent possible. This will maximize the number of sites that acquire concurrent total sulfur (e.g., SO₂ plus SO₄⁻) and total nitrogen (i.e., NO₂, NO₃, NH₃, and NH₄⁺). Special studies need to be conducted to compare measurement vs. model estimated dry deposition velocities.

Section 7.2.2.3 “Relating Deposition-related Effects to Air Quality Metrics” notes that oxidized nitrogen deposition in rural areas is mostly from HNO₃ and particulate nitrate rather than NO₂. Areas with higher NH₃ emissions also suggest inorganic nitrogen (e.g., NH₄⁺) contributions (Lines 24-27, Page 7-21). Better correlations are found between annual average PM_{2.5} and nitrogen deposition as compared to NO₂ vs nitrogen deposition (Pages 7-21 and 7-22).

Nitrate formation is associated with HNO₃ and NH₃, while HNO₃ formation depends on NO_x and volatile organic compounds (VOCs), similar to ozone formation (Stockwell et al., 2000). Additional research to assess limiting precursors for HNO₃ and particulate nitrate formation under current emission and meteorological conditions for sensitive ecoregions is needed. Both gaseous precursors and secondary inorganic aerosol need to be measured. Thermodynamic equilibrium model simulations (e.g., Simulating Composition of Atmospheric Particles at Equilibrium (SCAPE, Kim et al., 1993), ISORROPIA, Greek for "equilibrium", Nenes et al. (1998)) can be applied to better understand the different responses of PM_{2.5} mass and nitrate concentrations to changes in SO₄⁻, HNO₃, and NH₃ concentrations at different geological regions. The effectiveness of particulate nitrate reduction may depend upon interrelated NH₃ and NO_x reductions (e.g., Blanchard and Hidy, 2003, 2005; Blanchard et al., 2007).

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Dr. Charles T. Driscoll, Jr.

Overview

I appreciate the opportunity to serve on the CASAC review committee. In general, I found the document Policy Assessment for the Review of the Secondary National Ambient Air Quality Standards for Oxides of Nitrogen, Oxides of Sulfur and Particulate Matter, External Review Draft to be an interesting and informative document. I was challenged to review the document in the limited time provided and I received the charge questions after I started the review. Nevertheless, in general the review was well done, effective and clear, but I have some concerns and suggestions. I provide my general comments on the PA, responses to each of the charge questions followed by specific comments, questions, and suggestions.

Recent literature

I have concern that the ISA developed for this PA is now somewhat dated. The PA does not include or consider recent relevant literature that could bolster or modify the EPA's thinking or argument on the need for a revised standard and if so the approach and justification for a new or revised standard. I find this situation to be problematic. This gap in literature between ISAs and PAs is always a challenge for EPA. But during this review that problem is amplified because of the extended period since the cut-off year of literature for the ISA and the considerable relevant scientific literature that has been published since that date. It appears that the EPA has attempted to mitigate this deficiency as there are a few recent references added to the ISA. Below are a few recent papers that I participated in that contain information that could be considered by the EPA in their standard setting process for NO_x, SO_x and PM. If these papers from only me and my associates that could be relevant to ecosystem effects, think about all the recent literature that has not been considered for this PA for a Review of the Secondary National Ambient Air Quality Standards for Oxides of Nitrogen, Oxides of Sulfur and Particulate Matter. While this situation is problematic for EPA, at this point in time I don't know what can be done about it.

- McDonnell, T. C., J. Phelan, A. F. Talhelm, B. J. Cosby, C. T. Driscoll, T. J. Sullivan and T. Greaver. 2023. Protection of terrestrial ecosystems in the Eastern United States from elevated atmospheric deposition of sulfur and nitrogen: A comparison of steady-state and dynamic model results. *Environmental Pollution*, 318,120887. doi:10.1016/j.envpol.2022.120887.
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Deposition based standards

I understand that the history of the NAAQS is that they have been concentration based. Nevertheless, ecosystem effects are largely manifested through atmospheric deposition as evidenced by the material presented in the ISA and this PA. The EPA goes through an extensive evaluation in chapter 6 of the PA of translations of deposition-based effects. Under section 109(b)(2) of the CAA, a secondary standard must “specify a level of air quality the attainment and maintenance of which, in the judgment of the Administrator, based on such criteria, is requisite to protect the public welfare from any known or anticipated adverse effects associated with the presence of [the] pollutant in the ambient air.” Based on this wording of the CAA I see no reason why a NAAQS couldn’t be deposition based. The wording “level of air quality” is sufficiently vague that it could include deposition. Having a deposition-based standard would be a cleaner, more defensible approach because ecosystem effects are largely characterized and quantified on the basis of deposition. Moreover, the translation of deposition-based effects to an ambient concentration is fraught with difficulties and complexities as articulated in chapter 6.

Nitrogen effects in terrestrial ecosystems

I disagree with the conclusion in chapter 5 of the PA that there is generally a lack of quantitative information of nutrient enrichment effects of N. I refer the EPA to the analysis of Pavlovic et al. (2022) in which machine learning was used to analyze the effects of atmospheric N and S deposition on the growth and survival of species of trees using data from the USFS Forest Inventory and Analysis (FIA). The machine learning approach provided a more robust analysis quantifying species level response of tree growth and survival to recent levels of atmospheric S and N deposition than had been previously determined for the coterminous U.S. Moreover, these investigators characterized uncertainty limits for tree species critical loads of N and S. Machine learning can also be used to characterize the mediating factors that influence the response of tree species atmospheric deposition. This and other recent analysis indicate that effects from atmospheric N and S are continuing for many trees species across the U.S. and can be characterized and quantified.

Steady State vs Dynamic CL models

Much of the analysis on aquatic and terrestrial effects of acidification is based on steady-state modeling. The text of the PA in chapter 5 should be expanded to include a description of the assumptions associated with steady state modeling and the limitations of the application of such models (e.g., see McDonnell et al. 2023). The PA mentions but does not elaborate on dynamic models. Dynamic models are an important tool. They can be used in tandem with steady state models to provide important perspective. There have been important efforts in dynamic modeling in recent years that could bolster information on effects on soil nutrient concentrations and pools and the time frame for watershed ecosystem recovery (see McDonnell et al. 2021, 2023; Shao et al. 2020, Fakhraei et al. 2016, 2017). The EPA might want to consider adding some text on dynamic modeling results in chapter that helps put in context the steady-state CL modeling that is an important component of the PA.

Form of the standard

I have mixed feelings about the proposed form of a potential NO_x-SO_x-PM secondary standard. On one hand I wish the EPA would propose a deposition-based standard. Ecosystem effects of oxides of nitrogen and oxides of sulfur are largely driven by atmospheric deposition. So a deposition-based standard would

be a cleaner, more rational and defensible approach to a secondary standard. However, I am intrigued by a PM based standard. The reason for this is that N deposition effects to ecosystems are due to total N deposition. A secondary standard for NO₂ would not address the effects of reduced N. But since NH₄⁺ contributes to PM, a PM based standard would partially address the ecosystem impacts of both reduced and oxidized N. The concern I have with a PM based standard is that for some areas of the U.S. elevated PM is due to factors other than S and N. Going forward as SO₂ and NO_x emissions continue to decrease and forest fires and suspension of soil associated with increases in aridity continue to increase, the contribution of non N and S PM will increase, making this form of the standard problematic in the future.

Conduct a careful check of the text of the PA. There are several small mistakes that should be corrected before the document is finalized.

Chapter 1 – Introduction

To what extent does the Panel find that the information in Chapter 1 is clearly presented and that it provides useful context for the review?

Chapter 1 is clear, complete and well organized.

Chapter 2 –Air Quality

To what extent does the Panel find that the information in Chapter 2 is clearly presented and that it provides useful context for the review?

Chapter 2 is very useful and provides great information to set up the review. I particularly like the visuals provided. I have a number of specific comments/suggestions below

Specific comments

2-2, line15, ... particles to form SO₄²⁻.

2-21 would it be possible to provide a visual illustrating results from the AMoN network?

Section 2-4 Would it be possible to clarify (define) what is meant by a “design value”?

2-29 Is it possible to provide a visual showing the contribution of NH₄⁺ to PM_{2.5}?

2-29, line 5. This values of 150 µg/m³ is a mistake, correct? It should be 35µg/m³.

2-44, last paragraph. Is the wording for this paragraph the same as the title for Figure 2-40? The paragraph indicates the deposition is ammonia, but the figure title indicates total reduced N. Which is it? The figure title wording is likely correct and if so the wording of the paragraph should be corrected accordingly.

2-45, lines 8 and 9 provide a citation for the natural sources of reduced and oxidized N.

2-47 Might provide a map indicating the regions with states for the Table 2-2 in text of appendix.

2-47 Table 2-2. It might also be useful to add columns indicating % contribution to U.S. deposition.

Chapter 3 – Current Standards and Approach

To what extent does the Panel find that the background information in sections 3.31 and 3.2 to be clearly presented and provide useful context for the review? What are the Panel's views on the general approach described in section 3.3 for considering the welfare effects evidence and the risk assessment information for review of the adequacy of the current NO₂, SO₂ and PM secondary standards?

Chapter 3 is generally clear and provides useful information. Some comments/suggestions for chapter 3 are provided below. I have some thoughts on the approach which I mentioned in the general comment above.

3-4, line 20. Should this be mixing ratio? Please define mixing ratio.

Chapter 4 –Nature of Welfare Effects

To what extent does the Panel find the summary of the current evidence on the ecosystem effects of oxides of nitrogen, oxides of sulfur and particulate matter in ambient air, including consideration of both the evidence for direct effects of the pollutants in ambient air and for the effects of the associated atmospheric deposition into aquatic and terrestrial ecosystems, to be technically sound and clearly communicated? What are the Panel's views on the consideration of the public welfare implications, in light of judgments and conclusions made in NAAQS reviews regarding effects on the public welfare?

In general, I found the summary on ecosystem effects of oxides of nitrogen, oxides of sulfur and particulate matter in ambient air to be adequate. I think the direct effects are straightforward and technically sound and clearly communicated. I have some concerns about effects of deposition which I mentioned in the general comments above. I have some specific comments/suggestions for chapter 4.

4-7, line 15 The text should be modified to "... elevated dissolved inorganic aluminum".

4-8 It probably would be useful to elaborate on the definition of ANC. Beyond the chemical definition ANC is determined by strong acid titration and values can be influenced by concentrations of naturally occurring organic acids.

4-14 second paragraph. The text is confusing in this paragraph on the nature of concentrations of base cations and aluminum. What is the matrix considered? Generally, this ratio is calculated for soil exchange sites but it could also be for soil solutions. The nature of BC and Al concentrations needs to be clarified in the text.

4-16, line 32 phosphorus

4-17, line 13 Should clarify that responses occur in N-limited waters.

Figure 4-2 In Nutrient enrichment box might clarify that this is for freshwater ecosystems.

Chapter 5 – Exposure Conditions Associated with Effects

Does the Panel find the presentation of information and quantitative analyses related to welfare effects of oxides of nitrogen, oxides of sulfur and particulate matter to be technically sound and clearly communicated? What are the Panel's views on the quantitative analysis of aquatic acidification risks, including the choice of analysis approach and spatial resolutions? Does the document appropriately characterize the results of the analysis, including key limitations and associated uncertainties?

In general, I found Chapter 5 technically sound and clearly communicated. I have some concerns which are addressed in the general comments above concerning the understatement of nitrogen effects in terrestrial ecosystems and the reliance exclusively on steady-state models to define aquatic acidification effects. I have some specific comments/suggestions on Chapter 5 indicated below.

5-2 paragraph 2. As discussed above I believe that the PA understates the state of our current quantitative understanding of terrestrial effects of nitrogen.

5-8, line 20 Give date for Baker and Schofield reference

5-9, line 6 Smoky

5-10, line 5 Brook trout is generally considered a relatively pH insensitive species, as indicated elsewhere in the text.

5-53, paragraph 2 As indicated for chapter 4 need to clarify the nature of Ca:Al and BC:Al. Are these expressed in terms of exchangeable cations or soil solution concentrations?

Chapter 6 – Relationships of Deposition to Air Quality Metrics

To what extent does the Panel find the investigation of relationships between air concentration and deposition to be technically sound and clearly communicated? What are the CASAC views on the consideration of key limitations and associated uncertainties?

I find Chapter 6 very interesting and useful. I don't agree with this section that a NAAQS need to be set in terms of ambient concentrations. It would be much more rational and defensible to establish a secondary standard in terms of deposition, which would eliminate the need to artificially translate deposition values in terms of ambient concentration as evidenced by the challenges presented in this chapter. See general comments above.

Chapter 7 – Review of the Standards

What are the Panel views on the discussion of key considerations in sections 7.1 and 7.2 and preliminary conclusions? What are the CASAC views regarding the areas for additional research identified in Chapters 3 and 4? Are there additional areas that should be highlighted?

I have a general comment on the form of the standard above. Under future research needs I have concerns about the future loss of air quality monitoring capability due to the elimination of sites from the CASTNET program. I also mention above about the potential for machine learning techniques to characterize and quantify air pollution effects and the mediating factors that influence these effects and increasing application of dynamic models to understand the time-dependent nature of ecosystem effects of atmospheric N and S. I have a few specific comments below.

7-20, line 25 numbers

7-30, line 2 Should mention that NH_4^+ deposition has increased.

References

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- McDonnell, T. C., C. T. Driscoll, T. J. Sullivan, D. A. Burns, B. P. Baldigo and S. Shao. 2021. Regional Target Loads of Atmospheric Nitrogen and Sulfur Deposition for the Protection of Stream and Watershed Soil Resources of the Adirondack Mountains, USA. *Environmental Pollution*, 281:117110. doi: 10.1016/j.envpol.2021.117110.
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Dr. Ivan Fernandez

Answers to Charge Questions

1. Chapter 1 is well written and informative. It provides a useful context for this review.
2. Chapter 2 is well written and informative. It provides a useful context for this review.
3. Sections 3.1 and 3.2 provide clear and useful background information on past reviews, including a description of the structure, function, and history of the AAI as it relates to the goals of the prior review. Section 3.3 appears to provide a logical framework for the assessment of the adequacy of the standards, consistent with the approach laid out in Figures 3.1 and 3.2.
4. The presentation of information and quantitative analysis is technically sound and clearly communicated. The linkage of nitrogen deposition to effects is understated and inconsistent with evidence from the most recent ISA. The discussion of public welfare considerations by the Administrator serves to educate the reader on the frame of reference for this decision process and the guidance available for this process through consideration of past judgements in this regard. Chapter 4 does a good job describing the state of knowledge regarding the impacts of atmospheric S, N, and PM on terrestrial and aquatic ecosystems overall with exception of nitrogen linkages as noted. Most deposition effects descriptions are well written and emphasize the strong connection well established in the literature between atmospheric concentrations, deposition, and effects. The chapter mentions that historical deposition complicates the evaluation of welfare effects under lower recent deposition rates but can do more to develop the importance of this complexity to the evaluation of future effects.
5. Chapter 5 builds on the presentation in Chapter 4 by highlighting critical evidence in support of the decision on standards having to do with priority pollutant exposures associated with welfare effects. This chapter is well written, generally clear although it can be dense with analytical details in sections, and successfully links the discussion of the scientific evidence to its policy objectives. Many of the general issues discussed here reflect other chapters including chapter 4.
 - a. This chapter builds on a thorough and comprehensive ISA that provided strong support for EPA's utilization of the best available science up until approximately 2017. My sense is that this significant time gap since the ISA has only a limited effect on the final recommendations, but critical recent literature should be included by EPA in the finalization of the PA.
 - b. Related to item a., I was uncertain about the EPA approach to the significant gap in time between the 2020 ISA (literature to ~2017) and the present. The draft PA notes that beyond the 2020 ISA EPA included "...additional policy-relevant quantitative air quality, exposure and risk analyses." It could be useful to further define the criteria used to determine if there was an attempt to update the ISA since essentially all the literature in the ISA is "policy-relevant"? Did EPA include more recent literature that further supported what was already known in 2017? Was only more recent key literature included that alters the scientific understanding of the ISA such that it could influence the Administrator's decision? In short, what was EPA's strategy regarding more recent literature?
 - c. The focus on aquatic acidification, largely based on modeling studies, the discussion of Critical Loads, the use of ANC as a key metric for the evaluation of acidification, and the

- emphasis on Sulfur in this analysis regarding aquatic acidification seem appropriate in light of the policy objectives of the discussion. The treatment of aquatic acidification is the most quantitative section of the analyses presented in this chapter. This body of research is extensive and has been developed over decades which has supported positive outcomes of ecosystem recovery. The PA appropriately highlights the evidence for these successes particularly relevant to sulfur deposition declines and improvements in surface water ANC. The full body of science around these processes of response and recovery are more complex as noted in panel comments, but the PA seems to adequately recognize that reality without describing that extensive literature which seems appropriate.
- d. The PA pays a lot of attention to the BC:Al ratio as the indicator for soil acidification and forest effects, particularly from modeling studies. The key reference supporting the BC:Al metric is from 1993 while more recent studies report on soil cation dynamics in both acidification and recovery using additional metrics. BC:Al in terrestrial ecosystems is not of parallel utility as ANC is in aquatic ecosystems, in my opinion, and the PA could point to other metrics noted in the recent literature.
 - e. The PA is to be commended for its efforts to describe the complex and challenging issue of nitrogen for both aquatic and terrestrial ecosystems. However, the PA fails to recognize the well-established linkages between nitrogen deposition and ecological effects which is well developed by comments elsewhere in this review. I think the PA would benefit from a concise overview in a few paragraphs of the complexity of nitrogen to include issues around reduced and oxidized forms; gaseous, dissolved and particulate phases; exposure vs deposition; both negative and positive impacts such as acidification vs enhanced growth; atmospheric vs runoff derived nitrogen; acidification vs eutrophication; freshwater vs estuarine effects; as well as the complex dynamics of nitrogen cycling over time, which is particularly relevant to ecosystem recovery. The reader learns about all of these but in various chapters and levels of detail, making the complexity even more challenging. Perhaps a single aggregation of these concepts as a point of reference early in the PA would assist the reader's understanding.
 - f. Two areas of consideration regarding the uncertainty in our current scientific knowledge seem understated in the draft PA.
 - i. The first is, as stated in the PA (5-51), that the "...extent of the influence of historical deposition...on the associations reported in these studies with metrics quantifying more recent deposition is generally not known." The concurrent 'recovery' from high historical deposition of sulfur and in most areas nitrogen with ongoing research on effects, challenges the certainty with which we 'know' about future ecosystem health based on past research. This deserves further emphasis.
 - ii. The second is that the PA makes only passing reference to the reality that our knowledge of S, N and PM impacts on ecosystems comes from past research, and the uncertainty around their impacts in the future is influenced by the accelerating changes to our climate system, including warming, intensification of hydrologic cycles, and rising atmospheric CO₂ among others. The PA refers to climate mostly in the context of historical climate as a factor considered in several studies referenced. There is an increasing literature on the intersection of a changing climate and other air pollutants. This has implications for our level of uncertainty

in our knowledge base. The PA can more clearly convey that the established science of past decades may not translate unaltered into the future. As noted in the CASAC review of the ISA, in this discussion the PA also should include clarifying statements on the differences between meteorological factors over specific time periods, long-term climate change, and changes in atmospheric chemistry, most notably CO₂ concentrations, relative to ecological response.

6. The investigation between air concentrations and deposition appears technically sound and clearly communicated. Key limitations and uncertainties appear carefully considered and discussed in this chapter.
7. Chapter 7 appropriately draws on the prior chapters to discuss key considerations relevant to the conclusions drawn in this chapter. Table 7-1 is a useful summation of key considerations and alternatives to consider by the Administrator. Additional research needs are consistent with the last PA and appropriate for the continued refinement of these N and S standards overall. While this PA makes passing reference to climate change or related environmental changes in the document, further emphasis on the importance of changing climate in the uncertainty around past research findings, and the need for an improved understanding of the implications of these changes in weather on future welfare risks associated with the current standards, are warranted.

Editorial Suggestions

PAGE	LINE	COMMENT
1-14	18-20	Awkward wording in this sentence. Can this be clarified
	21	‘of’ S and N compounds...
2-2	15	..to ‘form’...
2-3	2	The relationship with NH ₃ as noted here should be depicted as part of the conceptualization represented in Figure 2-1.
	29	The NH ₃ symbol should be introduced on line 2 for this page.
2-4	28	Replace ‘manmade’ with ‘anthropogenic’ to better link to the following sentence.
2-13	28-29	This sentence does not qualify which effects as primary effects might be more evident in urban areas, whereas secondary effects indeed focus on rural areas.
2-21	7	...CASTNET data ‘can’ still be very useful...
2-27	Fig 2-22/23	For readers with some common visual color deficiencies (e.g., red/green), the color for 0-25 ppb and 251-376 ppb in the legends are very difficult to differentiate. This applies to all figures with this palette.
2-42	7	Omit ‘)’
2-44	10	Omit ‘the’
2-45	Fig 2-40	Below the legend the label says ‘Pct of total N as reduced N 1921’. Should that be 2021?
2-50	27	Change to ‘...due to any revision...’
3-2	15	Change to ‘...concluded that they provide protection...’
3-5	7	Delete ‘in’
3-6	1	Should this bullet start with ‘Does...’?

3-12	21	Omit first 'and'
4-2	16	Perhaps begin the sentence with 'The active research on SO ₂ ...' to avoid the possible interpretation that the quality of the research has changed.
	29	'...in the U.S. <u>and</u> found that...'
4-4	27	Change 'timber' to 'forest' as timber might be understood to be just roundwood or lumber forest products and this really applies to trees in forests, regardless of the product intended (e.g., pulp, chips, pellets, biochar, nanocellulose, CLT, as well as lumber).
	33	'...N and S deposition <u>to</u> ecosystems...'
4-5	20	Change 'a hydrogen ion' to 'hydrogen ions are'
4-12	1	Here or later in the document the additional uncertainty due to limitations on our knowledge of the ecological consequences of atmospheric deposition of N and S and the interaction with accelerating changes in climate (e.g., warming, intensification of the hydrologic cycle) should be acknowledged.
4-13	26	Change '...where sugar maple does less well...' to "...where sugar maple is less well adapted..."
4-14	1	"...low weathering <u>rates</u> ,..."
4-16	13	To follow on my comment above about uncertainty, this paragraph on terrestrial uncertainty includes reference to climate, but seems inconsistent with the language used for the aquatic uncertainty discussion. It seems a clear statement of uncertainty about the implications of a changing climate on our understanding of the science around N and S deposition should be made and be consistent throughout the narrative.
4-24	30	"... <u>and</u> contain vegetation..."
4-25	13	Is CSS already defined earlier in the document?
5-1	6	"... <u>of</u> exposures..."
5-11	19-20	The first sentence is not a complete sentence.
5-14	23	Change 'is' to 'are' as the word "data" is plural. Indeed, throughout (e.g., line 26 on this page should be data 'were') the document this tense could be addressed.
5-16	12	...and national forest(s)...
	23	...ANC target(s)...
5-17	21	There is no close parenthesis for this sentence.
	22	"In (the) 2012 review..."
5-18	6	Change 'has' to 'have'
5-29	4	2018-20.
	Table 5-2	Why is the sequence of time periods in reverse order compared to Table 5-1?
	28	'...for an of the three...' Is there a word missing here?
5-30	3	'...ecoregion-time period(s) had more...'
5-37	2	2014-2016
5-44	18	Catchment(s)
5-45	32	The representation of nitrate varies in this paragraph (e.g. nitrate, nitrate (NO ₃ -)) and likely without an intended meaning. Make this consistent.

5-46	7	‘... <u>the</u> watershed mass-balance...’
5-47	17	Delete ‘a’
5-50	1-6	This para ends using ‘TN’ but began using ‘N’ on the previous page. If these are intended to be the same metric, then use the same symbol to avoid any possible uncertainty for the reader.
5-52	21-23	This sentence might be worded differently to clarify the meaning. Accelerated leaching of base cations is the <i>process</i> that can lead to 1. decreases in absolute concentrations of exchangeable base cations, 2. decrease in base saturation, and 3. decrease in Ca:Al ratio. As stated, it reads like three different metrics, not a process and two resulting metrics that reflect that process.
5-53	6	‘...and/or nutrient enrichment...’
5-56	Table 5-7	‘L’ should not be capitalized for aluminum in lower left box.
5-57	10	Something is missing ‘...S or N compounds at ???’
	29	Best to restate ‘effects’ on what, which presumably is the mortality end point noted at the beginning of the paragraph.
5-58	36	‘...with <u>the</u> metric...’
5-61	17	Change ‘categorized’ to ‘categories’
5-62	28	A general comment on the discussion of experimental N addition studies is that little attention is paid to the form and strategy of N treatments in comparing studies, which would add to the complexity of comparing study outcomes.
5-69	14	The ‘L’ in aluminum should not be capitalized.
5-70	13	Change ‘relatively’ to ‘relative’.
5-71	14	Delete ‘to’
5-72	22	Change ‘has’ to ‘have’
6-7	31	‘... <u>is</u> not measured...’
6-12	12	Delete ‘spatial’
6-20	29	‘.. <u>be</u> transported...’
6-22	29	‘...in each Ecoregion III area...’
6-25	Fig 6-14	The color palette for the data points is difficult to interpret for anyone with certain visual color deficiencies (e.g., red/green) or color blind.
6-31	4	Delete the ‘4’
6-41	8	As used here, ‘inorganic’ seems to suggest the other species mentioned are organic, and they are not. Is this intended to be ‘reduced’ nitrogen?
6-42	1	‘...of each <u>of</u> the analyses.’
7-2	13	Is there a word missing after ‘quantitative’?
7-3	8	‘... <u>and</u> include...’
7-4	31	‘...secondary <u>standard</u> .’
7-9	1	What about ‘..., in waters with ANC values below 20 ueq/L? Reads like the same implication for the below zero statement earlier in the sentence.
7-12	29	Perhaps say ‘...background <u>ambient</u> atmospheric <u>deposition</u> ...’ to make clear this is not referring to an experimental addition in any way.
7-14	34	Change ‘levels’ to ‘level’

7-17	9	I do not see a section '5.2.1.4' above broken out explicitly, only 5.2.1? Same in the TOC. I looked for it, because this statement using the phrase 'more recently' would benefit from specific time periods of reference. For example, in the northeast 'recent' deposition reflects a greater decline in S than in N, so the time period can be relevant.
7-19	3-4	Here or elsewhere, in light of the reference to the length of studies, it is useful to note that, by their nature, trees and forests are long-lived and the evaluation of deposition effects are complicated by this reality and the varied conditions of the environment over these long time periods, different from studies of aquatic biota and seedlings.
7-20	25	'numbers'?
7-22	7	As written, it implies the monitors are the source of the pollutant, rather than the source of the pollutant data.
7-25	11	'in ambient <u>air</u> '?
	21	Continue to spell out 'DV' as has been the style for clarity.
7-27	34	Delete 'aspect'
7-28	32	'...control <u>of</u> N deposition...'
7-32	2	UNCERTAINTIES AND ???
7-33	24	Change 'contributed' to 'contribute'
	34-36	Subscripts for NO ₃

Dr. Frank Gilliam

Chapter 4 –Nature of Welfare Effects

To what extent does the Panel find the summary of the current evidence on the ecosystem effects of oxides of nitrogen, oxides of sulfur and particulate matter in ambient air, including consideration of both the evidence for direct effects of the pollutants in ambient air and for the effects of the associated atmospheric deposition into aquatic and terrestrial ecosystems, to be technically sound and clearly communicated?

In general, I liked the organization of this chapter that added clarity and ease of understanding. It appropriately acknowledged that the Clean Air Act has been quite successful in decreasing emissions of oxides of S and N. The chapter cites work to suggest that there has been improvements in tree species (e.g., increased growth rates) in response to declines in SO_x. What is not included in this assessment is the unavoidable confoundment that during this same extended period of decline in SO_x there has also been unprecedented increases in CO₂. The study in WV took this into account, but in general it should be acknowledged that there is difficulty teasing out effects of decreased SO_x and increased CO₂.

Of course, later additions to the CAA were equally effective in limiting NO_x emissions. Although Stevens (2016) is an excellent review, up to that time, of what was known regarding recovery of N-impacted ecosystems to declines in N deposition, a far more relevant reference would be Gilliam et al. (2019). It develops a hysteretic model to predict future change in forest ecosystems of eastern North America (and actually has three members of this CASAC as authors).

Gilliam FS, DA Burns, CT Driscoll, SD Frey, GM Lovett, and SA Watmough. 2019. Decreased atmospheric nitrogen deposition in eastern North America: predicted responses of forest ecosystems. *Environmental Pollution* 244:560-574.

What are the Panel's views on the consideration of the public welfare implications, in light of judgments and conclusions made in NAAQS reviews regarding effects on the public welfare?

Specifically defining 'public welfare' seems innately difficult, so I think that this chapter does a decent job articulating public welfare implications, especially in the context of the NAAQS review. I especially found the conceptual model at the end to be an effective way of not only depicting public welfare implications, but also putting them in the context of the other topics covered in the chapter.

Chapter 5 – Exposure Conditions Associated with Effects

Does the Panel find the presentation of information and quantitative analyses related to welfare effects of oxides of nitrogen, oxides of sulfur and particulate matter to be technically sound and clearly communicated?

As with Chapter 4, this chapter is generally well-organized and clearly written. For the most part, it balances the discussion between exposure as atmospheric concentrations and deposition, which, as a

biogeochemist, I find more relevant in assessing effects. Also as with Chapter 4, the hysteretic predictions of Gilliam et al. (2019) can well inform this chapter.

On the other hand, I am a bit confused regarding types of studies cited, especially for effects of excess N. The chapter appears to prefer agricultural studies over empirical work in forest ecosystems. Numerous researchers have studied the effects of N added to an entire watershed at the Fernow Experimental Forest for ~30 years. This includes many of the effects ostensibly reviewed in this chapter, including herb community response, soil fertility/chemistry responses, and stream chemistry responses. If this document is supposed to represent the state of the science in these various areas, omitting these recent (2016 to the present) studies makes it fall short of this aim.

What are the Panel's views on the quantitative analysis of aquatic acidification risks, including the choice of analysis approach and spatial resolutions?

Although this is not within my area of expertise, I feel that this section is one of the strongest of the chapter. I especially like the development of the use of ANC as a predictive metric in assessing such risks.

Does the document appropriately characterize the results of the analysis, including key limitations and associated uncertainties?

Each chapter ends with a reasonable coverage of limitations and uncertainties, and this one is no exception. I repeat my observation of a lack of relevant studies cited as a comment on how appropriate the chapter characterizes the results of the analysis. Fernow is not the only omission. There have been excellent recent papers out of Bear Brook Watershed in Maine (most recent one cited in this chapter is 2003).

Dr. Daven Henze

Chapter 2 –Air Quality

To what extent does the Panel find that the information in Chapter 2 is clearly presented and that it provides useful context for the review?

The overview of SO_x/NO_x/PM sources, transport, and deposition is overall quite valuable and useful in the context of the review. I've some minor comments and questions detailed below that help clarify some aspects of the presentation. Outside of these, a larger overall question is why satellite remote sensing based assessments of trends and distributions of sources and concentrations of NO₂, SO₂, NH₃, and PM_{2.5} are not discussed more comprehensively (only NH₃ is mentioned). I understand that such observations are, at present, not accepted as FRM or even FEM. However, the information they provide on trends and spatial gradients does seem relevant, in particular for construction and evaluation of components of the Nr deposition budget within TDEP, parts of which (i.e., NH₃ dry dep) are based on CMAQ model estimates alone.

In a similar vein, it would be nice to see a bit more regarding NH₃ measurements and NH₄⁺ concentrations. I recognize NH₃ is not a criteria pollutant. However, NH₃ is an unescapable and critical component of understanding reactive nitrogen deposition. Figure 2-40 shows the pronounced role of reduced N deposition in a majority of the area of CONUS. The discussion of why PM_{2.5} is a good indicator of N dep (compared to aerosol nitrate) in Chapter 6 hinges upon the role of NH₃. Thus, at a minimum it would useful to see analysis of NH₃ trends such as is shown (Figs 2-31 – 2-34) for sulfate, nitrate, and PM, and inclusion of NH₄⁺ in evaluation of PM_{2.5} composition (Fig 2-26).

Specific comments on Ch 2:

2-2, Figure 2-1 and supporting text: The description of composition is a bit simplified. SO₄²⁻ isn't the only PM form of sulfur oxides. S(IV) exists as H₂SO₄(aq) + HSO₄⁻ + SO₄²⁻, dry aerosol can contain NH₄HSO₄ as well as (NH₄)₂SO₄, etc. The speciation can have important implications for understanding PM trends (e.g., Silvern et al., 2017). Perhaps it should be noted that “sulfate” is taken here to refer to all of these? Also, both nitrogen oxides and sulfate also lead to formation of OC in PM via oxidation to form SOA.

2-3, line 20: N₂O₅ hydrolysis is the main pathway in winter — seems it warrants a more explicit explanations than “via reactions with other oxidants and water.”

2-3, line 21: “Under the right thermodynamic conditions” is also rather vague. It's easy enough to say that cold, humid conditions help, along with availability of excess NH₃.

2-3, line 28: Reduced N is distinct from oxidized N, that's true, but this statement reads as if only the former contributes to PM_{2.5} and adverse deposition-related effects.

2-5, lines 5-6: This could more explicitly say that inventories are thus uncertain, and even state estimated uncertainty ranges for different sources (e.g., SO₂ from power plants <20%, whereas NO_x from transportation up to 50%, NH₃ from all sources up to 100%...). These are mentioned for NH₃ on

page 2-12, which is good. Perhaps some discussion for uncertainties in sources of other species would be good to include as well.

2-6/Fig 2-2: I was a little surprised that the NEI contains burning and soil emissions. Were these generated by other models? For example, soil NO_x in CMAQ from modules like BDSNP or Rasool et al. (2019). Likewise, burning emissions are typically from inventories such as GFED or FINN.

2-7/Fig 2-4: Regarding NO_x trends, there are several studies discussing these trends and suggesting that more recent decreases may be less severe. Also, no impacts of COVID-19 lockdown are shown for spring 2020, even though these were known to significantly decrease transportation NO_x emissions (e.g., Harkins et al., 2021).

2-10: Line 4/5 states that NO_2 isn't directly emitted, which I don't believe is correct (though it is emitted both as NO and NO_2 , and rapidly interchanges). For example, O'Discoll et al. (2016) report PEMS measurements of vehicle emissions for a set of diesel vehicles whose NO_x emissions were 44% NO_2 .

2-14: The section on NO, NO_2 , and NO_x measurements is a bit jumbled and unclear. First, "direct measurement methods" is vague. Second, the "two step" process isn't clear (measurement of NO is mentioned, but how about NO_x ?). Next, the following sentence confusingly states that chemiluminescent measurements detect NO, NO_2 , and NO_x , though the previous sentence said they don't detect NO_2 . Further, the next few sentences discuss measurement biases, but it isn't clear how these are connected back to the previous issues of detecting only NO or NO_x (where the latter is actually going to be NO_y). Lastly, it is noted on 2-40 that species such as HONO and N_2O_5 have concentrations that are largest near sources and in urban areas, somewhat in contrast to 2-14, line 9, which states that urban concentrations of interfering species are relatively small.

2-16/17: It seems PM_{10} measurements should be mentioned somewhere, either within 2.3.3 ($\text{PM}_{2.5}$ measurements) or 2.3.4 (other).

2-29: Figure 2-26: Why not include NH_4^+ from NCore and CSN sites? These could perhaps be distinguished in some way from the IMPROVE sites, which don't have NH_4^+ ?

2-37/38: The differences between the process shown for constructing the TDEP dry dep between Fig 2-36 and 2-37 are a bit distracting. Why not make one figure with the detailed version? Also, the detailed version omits "other species" as estimated by CMAQ.

2-40, line 30-32: Regarding the reason for not bias-correcting TDEP NH_3 -- NO_x is also highly spatially variable, perhaps as much as NH_3 . Thus, I think it would be more precise to say here something like owing to the relative paucity of NH_3 observations.

2-41, line 5: The example on model biases for dry and wet deposition having a compensating effect when considering total deposition is true, but doesn't seem particularly relevant given that the model (CMAQ) is used only for bias correction of the dry portion of the TDEP estimate, whereas the wet portion is determined empirically (Fig 2-36). Thus, wouldn't a high bias for dry dep velocities lead to a high bias in total deposition?

2-45, lines 8-10: NO_x is also from natural sources like fires and lightening; natural NH_3 is also from fires.

2-45: It's not clear what is meant by relatively short lifetime (hours? days?), but in this context it seems important to distinguish between the gas-phase lifetime (hrs) and the particulate phase lifetime (days).

2-50 and Fig 2-43: Were NH_3 emissions held constant for these simulations, or, more realistically, did they continue with a slight increasing trend?

Chapter 6 – Relationships of Deposition to Air Quality Metrics

To what extent does the Panel find the investigation of relationships between air concentration and deposition to be technically sound and clearly communicated? What are the CASAC views on the consideration of key limitations and associated uncertainties?

Overall, there is a need to introduce more stringent standards, as indicated by the administrator's prior judgement that current standards do not adequately provide protection against adverse deposition-related effects. The coupling to the primary $\text{PM}_{2.5}$ standard of 12 ug/m^3 would be a tightening, while conveniently sustaining the practice of mitigation of welfare effects through primary standards. A concern is that reliance on $\text{PM}_{2.5}$ concentrations as an indicator could lead to situations where the "wrong part" of the $\text{PM}_{2.5}$ is reduced to attain the primary standard, from the perspective of reducing N deposition, especially as reduced nitrogen is an increasingly large component of N deposition yet NH_3 emissions are not regulated. Scientifically, it would be more direct to target N deposition values themselves as an indicator, which is perhaps permissible within the CAA's definition of a "level of air quality." That being said, below are my specific comments with regards to this question and the reviews approach to quantifying the relationships between air concentrations and deposition.

6-7, lines 25-33: Why not separately consider wet oxidized and reduced N (rather than total) deposition vs IMPROVE sulfate, nitrate and $\text{PM}_{2.5}$? This could help explain why they don't find a robust relationship between nitrate and total wet N deposition.

Figs 6-4, 6-5, 6-9: Overall I find these matrices very interesting and helpful, but they also take a while to absorb and are tricky to compare. In particular, they could be improved to help support the use of $\text{PM}_{2.5}$ as an indicator for N dep. Here are a few things that would help:

- Similar to my comment above for 6-7, evaluating oxidized vs reduced N deposition would be useful.
- The modeled vs measured matrices would be much easier to compare if ordered similarly, including the same quantities first (i.e., S + N deposition, N, NH_4^+ ,...). The same even applies just between Figs 6-4 and 6-5 (where e.g. the order of NADP NH_4^+ and NO_3^- are swapped).
- There is a lot of variability in measured N dep not explained by aerosol nitrate or nitric acid alone; the clear omission is NH_x . Can comparison to AMoN NH_3 be considered from measurements?
- To help address the previous point, a useful addition to the model matrix (Fig 6-9) would be NH_3 (g) and aerosol NH_4^+ , and for consistency with the measured matrices it would also be good to include $\text{PM}_{2.5}$. This is important as the combination of these would allow us to better understand the role that ammonia plays on $\text{PM}_{2.5}$ as an indicator of N deposition, which contributes to the suggested use of $\text{PM}_{2.5}$ as a standard in this regard.

- There are several panels that seem to exhibit two distinct regimes (Fig 6-9) or bimodal relationships (6-4, 6-5) — it would be interesting to examine the data more, perhaps by location, to evaluate why these occur. It does seem that Fig 6-12 and 6-13 address the former perhaps, showing the time dependence may lead to the different clusters.
- The scales are quite different for NADP S (up to 50) vs CMAQ wet dep S (up to 12) — what is the reason for this? Is the model low by more than a factor of two? In contrast, NADP N and Total Dep N from CMAQ are the same scale.

Regarding the definition and use of EAQM: I appreciate the approach, and the review well demonstrates the value compared to just considering concentration levels in an eco-region, in terms of an air concentration indicator that is better correlated to deposition. That being said, I have several questions and concerns:

- The threshold of 1% for a site being considered representative seems low. The time of 48 hrs also seems low, given the role of secondary species and PM with lifetimes of 2-10 days. These issues are later recognized in discussion of uncertainties in 6-42. However, actual sensitivity studies should be presented to evaluate the robustness of the relationships between deposition and EAQM values with regards to these assumptions.
- The EAQM approach based on HYSPLIT (meteorological trajectories that neglect chemistry) could miss situations where an emission of e.g. SO_x or NO_x in one location could reduce deposition at a location downwind (by promoting secondary PM formation that travels further than the target area). We've shown the role of chemistry in such situation in our previous work evaluating source contributions to reactive nitrogen deposition in biodiversity hotspots (Paulot et al., 2013) and Federal Class I regions (Lee et al., 2016). I'm not saying that this type of highly non-linear behavior dominates, this is an extreme case, but I do think that in general, chemistry and thermodynamics shouldn't be neglected when considering air / deposition relationships.
- A lot of Nr transport and dep will be seasonable, because of the seasonal nature of nitrate formation and NH₃ sources. The EAQM definitions based on transport patterns over the entire year may not be indicative of the transport patterns during the "deposition season." This would be another thing to consider via sensitivity calculations.

Fig 6-14, 15, 17 and 18 — it seems the relationship is very weak for the most recent two time periods. While this approach may work historically over the past two decades, it's not clear that below S dep levels of e.g. 5 kg S/ha yr there is a strong overall relationship across all sites. The text though does mention this in several places. However, I'd also note that placing the statistics (r) referred to in the text directly on the plots would be useful.

Fig 6-25: This is a rather remarkable figure, and to be honest I'm somewhat skeptical regarding the lack of scatter. I understand that PM_{2.5} will capture both reduced and oxidized N. If that's the explanation for this very high correlation, it could have been tested in earlier sections by considering more evaluation with NH₃, NH_x, and oxidized vs reduced N deposition. Regardless, there is still a large fraction of PM_{2.5} mass that is not associated with N of any kind (as shown in Fig 6-24), which makes the very tight correlations shown here difficult to believe. It is also concerning, in terms of a standard, for cases where PM_{2.5} exceedences above 12 ug/m³ are driven by non S or N rich sources, such as carbonaceous PM from wildfires.

Following receipt of corrections to Ch 6:

The differences between the original and corrected analysis of statistical relationships between EAQM NO₂, PM_{2.5} and N deposition are very different, and quite significant given the use of these relationships for developing the policy options in Chapter 7. For example, the correlation for PM_{2.5} shown in Fig 6-25 has reduced from 0.98 to 0.52. The correlations for NO₂ shown in Figures 6.20 and 6.21 have all but gone away. The figures also show considerably more bifurcation / clustering.

Such significant differences compared to that presented originally makes me further concerned about the EAQM analysis. It is a new analytical approach, which to my knowledge has not yet been peer reviewed. I'm concerned there may be other mistakes or corrections, or surprises in the relationships noted. There is long list of questions related to uncertainties and sensitivities listed above. I believe these should be addressed and the methods fully explained via a peer review process prior to forming a basis for the PA. I recognize that the PA (or RIA, in the past) has included non-peer reviewed results. However, in the past this has been through application of well-documented methods (e.g., SMAT) that depend upon relative response factors modeled using e.g. CMAQ under different emissions scenarios. In other words, this has been conducted using established, peer-reviewed methods. This seems to me to be an important distinction compared to EAQM. CASAC can serve as the peer review for new methods, if we are given ample time to request additional details, and review a revised PA, like we would with a standard journal article. A concern though is that the timeline doesn't necessarily permit this level of iteration.

Some specific comments on the corrected version:

6-32*, line 16: A change in r from 0.52 to 0.03 is more severe than diminished — there is no significant correlation in the latter.

6-32*, line 22: This is somewhat subjective, but I would say a correlation of 0.12 is not just less than 0.63, but essentially shows little to no correlation.

Figures 6.20, 6.21, 6.25, 6.26: These figures clearly show bifurcation / clustering in the relationships between N deposition and EAQM NO₂ and PM_{2.5} concentrations. I recognize that the EPA memo communicating the corrections to Chapter 6 presents explanations for these bifurcations in terms of spatial differences (Eastern vs Western ecoregions). These explanations and supporting figures and tables should be included within Chapter 6 itself, which at present in the section containing these figures does not even contain the words “eastern” or “western”. It would further be useful to see if correlation of N deposition by oxidized vs reduced N lead to significant adjustments to correlations with EAQM NO₂ and PM_{2.5}, or if there are other explanatory factors.

Additionally, it would be very useful to support recommendations in Chapter 7 if EPA could provide:

- Statistics broken down by all data vs only considering data from the two most recent time periods, given the latter are most relevant in terms of ambient chemical conditions that impact the lifetimes and compositions of the species of concern
- Statistical significance (p-values) of any correlations reported

*Track changes version, document NOxSOxPM-CASAC Memo-CorrectedPApages0727.pdf.

Editorial

1-9, 14: Typo — extra “10”

2-2: Note that section 2-3 begins with a quick overview of NO_x sources, where here the intro to SO_x does not discuss sources.

2-2/2-3: The following two sentences are repeated verbatim in two consecutive paragraphs: “Dry depiction is an important....Although particulate sulfate....”

2-3: Why is x of NO_x subscript but x of SO_x is not? Seems like it should be, for both. NH_x was also given a subscript x.

2-19: For Fig 2-15, the “dropped pin” marker isn’t the greatest, as it’s hard to tell where many pins touch. I’d suggest a plot more like Fig 2-14. The same applies to Fig 2-17, though to a lesser extent.

2-23: Does the 80 ppb on line 10 refer to the max or the actual design value, i.e., the 98th percentile? The text is a little unclear, but caption of Fig 2-18 says 98th percentile.

2-29, line 13: to be

2-42: proxy)

2-44: within in

2-44: the each

6-3, line 1: Near ground-level, rather than for ground-level?

6A-1: three separate pollutants

References

Harkins, C, B. C. McDonald, D. K. Henze, and C. Wiedinmyer (2021), A fuel-based method for updating mobile source emissions during the COVID-19 pandemic, *Environ. Res. Lett.*, 16, 065018, <http://dx.doi.org/10.1088/1748-9326/ac0660>.

Lee, H.-M., F. Paulot, D. K. Henze, K. Travis, D. J. Jacob, L. H. Pardo, B. Schichtel (2016), Sources of nitrogen deposition in Federal Class I areas in the US, *Atmos. Chem. Phys.*, 16, 525-540, doi:10.5194/acp-16-1-2016

O'Driscoll, R. S., H.M. ApSimon, T. Oxley and N. Molden, *Journal of Earth Sciences and Geotechnical Engineering*, vol.6, no. 4, 2016, 15-28.

Paulot, F., D. J. Jacob and D. K. Henze (2013), Sources and processes contributing to nitrogen deposition in biodiversity hotspots worldwide, *Environ. Sci. Technol.*, 47, 3226-3233, doi:10.1021/es3027727.

Silvern, R. F., Jacob, D. J., Kim, P. S., Marais, E. A., Turner, J. R., Campuzano-Jost, P., and Jimenez, J. L.: Inconsistency of ammonium–sulfate aerosol ratios with thermodynamic models in the eastern US: a possible role of organic aerosol, *Atmos. Chem. Phys.*, 17, 5107–5118, <https://doi.org/10.5194/acp-17-5107-2017>, 2017.

Rasool, Q. Z., Bash, J. O., and Cohan, D. S.: Mechanistic representation of soil nitrogen emissions in the Community Multiscale Air Quality (CMAQ) model v 5.1, *Geosci. Model Dev.*, 12, 849–878, <https://doi.org/10.5194/gmd-12-849-2019>, 2019

Dr. Robert W. Howarth

With regard to the charge to the Panel, I strongly disagree with the conclusion in Chapter 7 that the present standards are adequately protective. The largest failing is the lack of consideration of NH₃ gas, despite the role of NH₃ in fine particles and the adverse consequences of ammonia on both aquatic and terrestrial ecosystems (including eutrophication of estuaries and coastal marine waters, toxicity of cyanobacteria blooms (HABs), and acidification of freshwaters and soils). The standards also do not adequately address vehicles as a source of both NH₃ and NO_x that contribute to coastal eutrophication.

The major problems with the overall report that I see include:

1) Inadequate consideration of eutrophication of estuaries and coastal marine waters:

The draft PA has astoundingly little discussion or analysis on the role of nitrogen deposition on water quality in estuaries and coastal marine waters. Numerous studies, reports, and peer-reviewed papers have identified nitrogen pollution and the resulting eutrophication of coastal waters as one of the greatest threats to the estuaries and coastal waters of the US. None of this is captured in the draft PA, which has a total of only 31 lines – less than one page -- that even mention estuaries and coastal marine ecosystems (page 4-17, lines 6-9; page 4-18, lines 14-16; page 4-19, lines 3-5 ; page 4-20, lines 16-19 ; page 4-20, lines 24-27 ; page 4-21, lines 6-8 ; page 4-21, lines 11-13 ; page 5-48, lines 13-15 ; page 7-17 bottom, to top of page 7-18). The draft PA actually spends more time discussing nitrogen as a cause of eutrophication in freshwater lakes and wetlands, although the case of nitrogen causing eutrophication in lakes is much weaker than for estuaries: most eutrophic lakes are limited by phosphorus in the US, while eutrophic estuaries are nitrogen limited.

On page 5-48, the draft PA states “With regard to the effects related to N enrichment in various types of aquatic ecosystems, such analyses were not performed due to recognition of a number of factors, including modeling and assessment complexities, and site- or waterbody-specific data requirements, as well as, in some cases, issues of apportionment of atmospheric sources separate from other influential sources.” With this justification, the effects of nitrogen in estuaries and coastal marine ecosystems receive no further discussion. I find this to be unacceptable, given the societal interest in estuaries, the proven role of nitrogen as a leading cause of degradation in estuaries, and the very large literature on sources of nitrogen (including atmospheric deposition) to estuaries and the consequences of this nitrogen.

2) Inadequate analysis on the role of atmospheric deposition as a contributor to the nitrogen load to estuaries:

The draft PA has rather little discussion on the contribution of atmospheric deposition of nitrogen to estuaries, and in several places downplays this with statement such as it is difficult to estimate how much of the nitrogen load to estuaries comes from atmospheric sources (see for example page 7-28). However, at one point the draft PA indicates that atmospheric deposition can contribute 15% to 40% of the total nitrogen load to estuaries (page 4-20). To me, this seems quite substantial, given the huge problem of nitrogen leading to eutrophication in coastal marine ecosystems. On this basis alone, the draft PA really needs to expand this presentation and discussion. Further, I would note that several lines of evidence suggest this is an underestimate (see for example my 2008 paper on this, Howarth, R. W. 2008. How important is atmospheric deposition as a source of nitrogen to coastal marine ecosystems in

the northeastern United States? Pages 47-65 in A. Desbonnet and B. A Cost-Pierce (eds.), *Science of Ecosystem-Based Management*. Springer, NY). And all of the studies in my 2008 paper as well as I believe those behind the 15% to 40% estimate made on page 4-20 are only for deposition of oxidized forms for nitrogen. Deposition of reduced forms of nitrogen may also substantially contribute to nitrogen loads to estuaries, with resulting eutrophication. Regarding the role of deposition of oxidized nitrogen as an input to estuaries, the NANI (Net Anthropogenic Nitrogen Inputs) model provides a comprehensive and systematic approach for all watersheds in the United States.

3) Inadequate consideration of vehicles as a source of NH₃ emissions:

The draft PA briefly mentions vehicles as a source of NH₃ emissions (page 2-3, lines 28-32, and page 2-10, lines 4-12), but there is very little presentation on this beyond the brief statement that “While mobile source contributions to total NH₃ emissions are only about 2% at the national level, there is a growing body of evidence suggesting that vehicular sources may be underestimated in the NEI (Sun et al., 2017; Chen et al., 2022). Any underestimation in mobile source NH₃ emissions would mostly impact urban areas, where there is a lot of on-road mobile source traffic.” Some literature suggests that catalytic converters may over reduce NO_x, producing NH₃ from car exhaust. Similarly, the use of urea to treat diesel exhaust from trucks may lead to significant NH₃ emissions. And there is some suggestion that NH₃ emissions from vehicles in urban and suburban areas may contribute significantly to the nitrogen load to Chesapeake Bay (Howarth 2008 paper cited above). Further, our research on Cape Cod indicates that vehicle exhaust may contribute up to 25% of the total nitrogen load to a eutrophic, with the contribution by NH₃ greater than that from NO_x (Bettez, N., R. Marino, R.W. Howarth, and E.A. Davidson. 2013. Roads as nitrogen deposition hot spots. *Biogeochemistry* 114: 149-163).

4) Complete lack of consideration of the role of nitrogen in contributing to toxicity of cyanobacterial blooms in freshwater lakes:

The draft PA has some small discussion on nitrogen as a limiting nutrient in freshwater lakes (page 4-4, lines 1-3, and page 4-16, lines 21-22, for example). This is a relatively minor issue, and most eutrophic freshwater lakes in the US are limited by phosphorus, or sometimes co-limited by phosphorus and nitrogen. A bigger concern with nitrogen in freshwater lakes is the association with the toxicity of cyanobacteria blooms (HABs). These toxins pose a large public health threat to millions of Americans, and greater toxicity of blooms may also make blooms more likely, or more persistent, by reducing grazing on the cyanobacteria by zooplankton and benthic animals. Cyanobacterial toxins such as microcystin are very nitrogen rich, and higher concentrations of nitrogen in lake water can result in higher concentrations of the toxins.

Note that this is not related to eutrophication: high nitrogen levels can lead to high toxins levels even in oligotrophic, non eutrophied lakes. For example, Skaneateles Lake (NY) is quite oligotrophic, is strongly limited by phosphorus (not nitrogen), and is considered by the EPA to be one of the “cleanest” lakes in the US. Since 2017, however, the lake has had persistent toxic cyanobacterial blooms each summer, and the high nitrogen levels in the lake seem likely to contribute to this. Much of this nitrogen comes from atmospheric deposition (of both oxidized and reduced N).

5) Inconsistent and misleading presentation on trends in NH₃ emissions and deposition:

On page 2-12, the draft PA states that NH₃ emissions nationally have remained “relatively flat,” although it acknowledges substantial uncertainty in emission factors for NH₃. Later on page 2-34, the

draft states “Consistent with expectations from the flat or slightly increasing trends in ammonia emissions, we also see relatively unchanged NH₃ concentrations over this 10 year period, although there can be some variability from site to site.” Both of these statements fly in the face of a substantial body of evidence that shows an increase in deposition of NH₃ and NH₄⁺ over time, as for example at the CASTnet site run out of my lab (Butler, T., R. Marino, D. Schwede, R. Howarth, J. Sparks, and K. Sparks. 2015. Atmospheric ammonia measurements at low concentration sites in the northeastern USA: implications for total nitrogen deposition and comparison with CMAQ estimates. *Biogeochemistry* 122: 191-210). This is acknowledged later in the draft PA (see page 2-46), where it is stated that atmospheric NH₃ concentrations have risen over time, as has the deposition of reduced forms of N, in contrast to decreasing deposition of NO_y. However, the draft PA does not acknowledge the inconsistency with the earlier statements. One explanation for the contradiction between estimated emissions and measured deposition is that the emissions estimates are incorrect, and do not capture changes over time.

On page 2-41, the draft PA states “it is likely that the resulting estimates for current conditions reported in this assessment overestimate ammonia dry deposition due to the overestimate in ammonia concentrations. This error is most pronounced in regions near large ammonia emission sources, such as confined animal feeding operations (CAFOs) and fertilized crops.” I disagree, and note that we have previously demonstrated that CMAQ under-estimates (not over-estimates) dry NH₃ deposition at the CASTNet site managed from our lab, and that the under-estimation by the CMAQ model is even greater as one measured deposition in closer proximity to a CAFO (Butler et al. 2015, cited above). We have also demonstrated that CMAQ and other EPA models underestimate the dry deposition of ammonia that originates from CAFOs onto the watershed and lake surface of Skaneateles Lake, NY (Butler, T., R.W. Howarth, D.P. Swaney, C.B. Bablitz, and G.M. Beachley. 2023. Atmospheric ammonia deposition: An important source of nitrogen to an oligotrophic lake watershed, presented at the ASLO Aquatic Sciences Meeting, Mallorca, Spain).

Dr. Donna Kenski

Thank you for the opportunity to comment on the Policy Assessment for the Review of the Secondary NAAQS for NO_x, SO_x, and PM. Overall, the document was well written and concise, and generally presented at an appropriate level to be read and understood by interested parties. In a few areas the prose gets tangled up in itself and needs to be edited or rewritten for clarity; I have noted specific places in my comments below. A table of acronyms would be a welcome addition.

Chapter 1 – Introduction

To what extent does the Panel find that the information in Chapter 1 is clearly presented and that it provides useful context for the review?

This chapter provided a concise summary of the past standards and review processes and is indeed useful context for this review. I have no recommendations for changes.

Chapter 2 –Air Quality

To what extent does the Panel find that the information in Chapter 2 is clearly presented and that it provides useful context for the review?

This chapter was also clearly presented and provides useful context for the review. The maps and figures are well done and some are very enlightening. I noted with dismay that the area of highest ammonia emissions (Iowa, southern MN) has no ammonia monitors. Ammonia seems to remain an area of greater uncertainty and differing interpretations. Where EPA sees relatively unchanged ammonia concentrations (Sec. 2.4.4, Fig. 2-35), I see a significant increase, most markedly in the Midwest and Plains states. Some minor formatting issues remain, particularly inconsistent capitalization (Near-road, near-road), subscripts (So_x vs SO_x), and spelling (collocation, co-location).

Chapter 3 – Current Standards and Approach

To what extent does the Panel find that the background information in sections 3.3.1 and 3.2 is clearly presented and provide useful context for the review? What are the Panel's views on the general approach described in section 3.3 for considering the welfare effects evidence and the risk assessment information for review of the adequacy of the current NO₂, SO₂ and PM secondary standards?

This chapter was straightforward and clear. I have no recommendations for changes.

Chapter 4 –Nature of Welfare Effects

To what extent does the Panel find the summary of the current evidence on the ecosystem effects of oxides of nitrogen, oxides of sulfur and particulate matter in ambient air, including consideration of both the evidence for direct effects of the pollutants in ambient air and for the effects of the associated

atmospheric deposition into aquatic and terrestrial ecosystems, to be technically sound and clearly communicated? What are the Panel's views on the consideration of the public welfare implications, in light of judgments and conclusions made in NAAQS reviews regarding effects on the public welfare?

This chapter is not my area of expertise but I thought it was generally clear and concise. In particular Fig. 4-3 was a helpful schematic.

Chapter 5 – Exposure Conditions Associated with Effects

Does the Panel find the presentation of information and quantitative analyses related to welfare effects of oxides of nitrogen, oxides of sulfur and particulate matter to be technically sound and clearly communicated? What are the Panel's views on the quantitative analysis of aquatic acidification risks, including the choice of analysis approach and spatial resolutions? Does the document appropriately characterize the results of the analysis, including key limitations and associated uncertainties?

No comments.

Chapter 6 – Relationships of Deposition to Air Quality Metrics

To what extent does the Panel find the investigation of relationships between air concentration and deposition to be technically sound and clearly communicated? What are the CASAC views on the consideration of key limitations and associated uncertainties?

This was an interesting analysis that provides a lot of supporting evidence for the approach used to justify Chapter 7 and the accompanying policy decisions. Some of the assumptions and plots were not adequately addressed. Page 6-4, lines 15-17, declares that the assessment in Sec 6.2.1.1 (the collocated site analysis) relies on wet deposition as a proxy for total deposition since dry deposition is not routinely measured. The relatively high correlations seen in the scatterplots (Figs 6.4, 6.5, 6.10, 6.11) seem to imply that the assumption is valid, but there are some troubling bifurcations in the scatterplots that are not addressed in the text. The explanations on p. 6-7, lines 27-33, are all plausible but the sharp divisions seen in the plots don't look like random variability. Was there any attempt to tease out the differences? The same issue shows up in Figs. 6-23, 6-24, and 6-29. I appreciate the explanation given about non-sulfate PM_{2.5}, but some additional exploration of these data is warranted. Are some specific sites driving these patterns, perhaps?

The most interesting part of this chapter is Fig. 6-25, which is startling. For ambient data over that long period to correlate so closely with a modeled deposition value is extraordinary. I trust you have checked and rechecked this data? I don't have a good enough feel for the EAQM process and TDEP procedures to imagine how this could happen, but my skeptical self thinks it can't be real. If it is, though, it's amazing and certainly supports the novel idea that a PM concentration standard could conceivably control N deposition. I would, however, need that decision to be based on more than this one plot.

The EAQM model was poorly documented, and even Appendix 6A didn't give any significant details. A much more comprehensive discussion of the methodology is warranted, especially as it provides some critical plots informing the level of the standard.

Chapter 7 – Review of the Standards

What are the Panel views on the discussion of key considerations in sections 7.1 and 7.2 and preliminary conclusions? What are the CASAC views regarding the areas for additional research identified in Chapters 3 and 4? Are there additional areas that should be highlighted?

This chapter was more dense than most of the rest of the document and it could benefit from a little more editing. The information was comprehensive but not necessarily convincing, at least for the SO₂ standard. The ‘not to be exceeded’ form of the standard was not shown to be as protective as an annual average for deposition. It would be fine to retain the existing secondary standard or preferably tighten it for protecting from short term exposures, but the PA clearly points out that deposition by nature is cumulative over time. I believe the data support the need for an *additional* annual average standard, and based on the uncertainties outlined throughout the document, adopting an annual average standard in the range of 10-22 ppb seems reasonable, especially since it’s already attained almost everywhere. On p. 6-22 lines 19-20, EPA says ‘an annual standard is more relevant to assessing accumulated deposition than a standard with a form set to reduce peak concentrations.’ It seems only proper to make the standard fit the nature of the problem it’s trying to solve, rather than coopting an existing form. If EPA goes ahead with the not-to-be-exceeded form, they should demonstrate that it truly is protective for deposition. The current PA does not address that adequately. Also it would need to address the question of whether the secondary standard can be more restrictive than the primary standard.

There is a similar shortcoming for the proposed NO₂ and PM_{2.5} standards. Since NO₂ attainment has been achieved nationally for years, the real issue seems to be preventing backsliding and maintaining current conditions as maximums. The PA should be more transparent about this. Likewise for PM_{2.5}; this was the most interesting part of the PA. The notion that PM_{2.5} standards alone might be sufficient for controlling N and S deposition was new and exciting. The relatively simple correlations and EAQM analysis presented in this document were pretty compelling, but I think it requires more analysis if PM_{2.5} alone were to become the standard for controlling N and S deposition.

The discussion of the proposed standards would also be helped by a more direct discussion tying the levels directly to deposition. Of the hundreds (!) of plots in Chapter 6, show us the ones that tie PM_{2.5} to a deposition of XX kg/ha, and put a big red dot at the intersection of 12 ug/m³ and XX kg/ha. It seems obvious, and the data are there, but Table 7-1 required a lot of searching back thru Chapters 6 and 5 to find the supporting information.

At the CASAC June meeting several members mentioned the potential advantages of standards for particle nitrogen (NO₃ + NH₄) and/or particle sulfate, which were interesting options that I would like to see EPA explore, since they would more directly get at the species of interest.

The first full paragraph on p. 7-10 is really tough to parse. Please break this up into something more reader-friendly. I’m not sure that this level of quantification is necessary in this part of the document since it is covered in Chapter 5.

The last paragraph on p. 7-30 is interesting. EPA acknowledges the potential value of an alternate indicator, including NH₄, but declares the information gaps too daunting. I agree that we need more information, of course. But this document again sweeps NH₃ under the rug. Emissions are increasing, unlike for NO₂ and SO₂, and it has a host of direct and indirect effects on the environment. Please keep

it at the top of your research agenda. It seems inevitable that it will become more important if emission trends continue.

My views regarding additional research mostly concern funding. Future research will by necessity depend heavily on the data from our various national monitoring networks: NAMS/SLAMS, CSN, IMPROVE, CASTNET, NADP, etc. Nevertheless, all these networks are faced perennially with uncertain funding and cutbacks that threaten the continuity of the critical data stream that they provide. I repeat from my previous comments on the ISA and similar statements from multiple previous CASAC reviews: “Much of the data analysis presented in the ISA hinges on NADP, CASTNET, and IMPROVE data. These networks are partially funded by EPA, but each has a significant fraction of sites funded by non-EPA partners (some federal and some not). The continuity of each of these networks is constantly in peril due to funding uncertainty. Of course, assessing changes over time requires long term monitoring and stable networks. EPA must prioritize network funding, for these and the other networks that come under its umbrella.” I would add that the need for monitors in rural areas (i.e., CASTNET, IMPROVE) is more critical than ever as we are challenged by climate change and the need to understand air pollution impacts in remote parts of the country as well as urban centers. Their long-term existence is essential to maintain the protection of public health, natural resources, and sustainable economic growth. EPA must prioritize network funding, for these and the other networks that come under its umbrella.

Minor editing, typos, etc:

Throughout the document: Please be consistent with the use of the Oxford comma. I recommend using it, based on the dense prose in this document, but it is a style decision. Nevertheless, the inconsistent application is distracting and just makes it harder for readers to focus. Similarly, the word ‘collocated’ is spelled and hyphenated in various ways. The use of subscripts in earlier chapters alternates between capitals and lowercase.

p. 2-1, line 3: nitrogen, oxides -> nitrogen and oxides

p. 2-2, line 15: to for -> to become

p. 2-21, line 7: data still be -> data are still

p. 2-23, line 16: in Los Angeles -> in the Los Angeles

p. 2-42, line 7: remove stray end paren.

p. 2-42, line 17: sentence starting ‘Another issue...’ is unclear, please reword

p. 2-50, line 27-28: due to the any revision -> due to the potential for revision

p. 3-9, line 5: for -> from

p. 3-11, line 23: put a period after review

p.6-2, line 17: larger, coarse -> larger coarse

p.6-2, line 18: faster than smaller -> faster than that of smaller

p. 6-7, line 20: Figure 6-3 -> Figure 6-4

p. 6-17, line 12: again -> against

p.6-20, line 28: can -> can be

p.6-40, line 16: is -> in

p. 6-40, line 27: to somewhat -> to a somewhat

p. 7-2, line 13: quantitative for -> quantitative information for

p. 7-5, line 18: evidence were -> evidence was

p. 7-5, line 25: which is does -> which is available does

p. 7-6, line 4: then the consideration of -> then consider

p. 7-9, line 8: events, that -> events that

p. 7-11, line 4: in -> is

p. 7-14, line 35: relate uncertainties -> relate to uncertainties (maybe? My best guess...)

p. 7-16, line 29: evidence relates to the ~~evidence describing~~

p. 7-18, lines 7-12: please fix this run-on sentence

p.7-19, line 27: associates -> associations

p. 7-20, line 25: umbers -> numbers

p. 7-20, line 31: using database -> using a database

p. 7-20, line 31: over 23-year -> over a 23-year

p. 7-20, line 32: for 26-year -> for a 26-year

p. 7-26, lines 7-10: this sentence is not clear as written. Maybe remove the 'and' in line 9? Better to rewrite it.

p. 7-26, lines 24-25: near large SO₂ sources. ~~to provide control of these large sources of SO₂.~~

p. 7-27, line 34: specific ~~aspect~~-aspects

p. 7-28, line 32: for control -> for controlling

p. 7-29, lines 27-32: this is an unwieldy sentence. Please rewrite.

Appendix 6.A

P.6A-1, line 13: ‘for four separate pollutants: NO₂, SO₂, and PM_{2.5}’ should be ‘for three separate pollutants: NO₂, SO₂, and PM_{2.5}’

P6A-2, line 6: ‘and then the all’ should be ‘and then all’

P6A-2, lines 12-13: what is the significance of the fact that 2016 was a transition between a positive Nino Index and a negative one? I don’t see how that makes it more representative – could you elaborate? Was there any more analysis performed to find a year with ‘average’ meteorology, or defining what that might mean?

Table 6A-4 should be in Appendix 5 – there’s no discussion or relevance to Appendix 6 or Chapter 6, as best I can tell.

Dr. William McDowell

Chapter 1 – Introduction

To what extent does the Panel find that the information in Chapter 1 is clearly presented and that it provides useful context for the review?

The overarching approach to uncertainty throughout the report is somewhat imbalanced. Rather than focusing on uncertainty alone, the highly certain outcomes and clear findings from past research and monitoring should also be highlighted. This chapter might also benefit from an explicit acknowledgement that past successes in reducing emissions may now lead to more uncertainty about links between air quality and ongoing welfare effects, as they are now more subtle (though still of concern) than in past decades. This “new” uncertainty should be addressed rigorously, and may require new approaches to measuring, quantifying and regulating air quality.

Chapter 4 –Nature of Welfare Effects

To what extent does the Panel find the summary of the current evidence on the ecosystem effects of oxides of nitrogen, oxides of sulfur and particulate matter in ambient air, including consideration of both the evidence for direct effects of the pollutants in ambient air and for the effects of the associated atmospheric deposition into aquatic and terrestrial ecosystems, to be technically sound and clearly communicated? What are the Panel’s views on the consideration of the public welfare implications, in light of judgments and conclusions made in NAAQS reviews regarding effects on the public welfare?

The chapter provides a good summary of current understanding (at the time of the ISA) of the effects of the oxides of nitrogen, oxides of sulfur and particulate matter on aquatic and terrestrial ecosystems. The chapter is technically sound and clearly communicated. This chapter suffers, however, from the lag time between the ISA and the PA. Much recent literature shows that N deposition is changing (both in N species and often in total deposition; e.g. Murray et al. 2022; Nitrogen wet deposition stoichiometry: the role of organic nitrogen, seasonality, and snow. Biogeochemistry 2022 Vol. 160 Issue 3 Pages 301-314 DOI: 10.1007/s10533-022-00966-0). Furthermore, dissolved organic N deposition is important and increasing at this particular site, and may be increasing more broadly.

Additional evidence is now available showing that watershed N deposition affects stream N dynamics in many watersheds around the world (Templer et al. 2022; Atmospheric deposition and precipitation are important predictors of inorganic nitrogen export to streams from forest and grassland watersheds: a large-scale data synthesis. Biogeochemistry 2022 Vol. 160 Issue 2 Pages 219-241 DOI: 10.1007/s10533-022-00951-7).

The ever-increasing role of organic acids/DOC as a factor that may affect the fate of N in terrestrial and aquatic environments has also become more evident in the last 7 years (e.g. Monteith et al. 2023; Long-term rise in riverine dissolved organic carbon concentration is predicted by electrolyte solubility theory. Science Advances doi:10.1126/sciadv.ade3491). Although changes in both dissolved organic carbon and dissolved organic nitrogen (DOC and DON) concentrations in streams have occurred over the last few decades (driven at least in part to changing deposition), the patterns in DOC and DON are not always consistent (Rodriguez-Cardona et al. 2021. Shifting stoichiometry: Long-term trends in stream-dissolved

organic matter reveal altered C:N ratios due to history of atmospheric acid deposition. Global Change Biology DOI: 10.1111/gcb.15965).

Specific comments for clarification:

Page 4-3 line 1-4: Further support for this conclusion regarding the importance of direct air quality effects on tree growth is provided for the Czech Republic in Hruska et al. 2023 (Hruška, J., F. Oulehle, T. Chuman, T. Kolář, M. Rybníček, M. Trnka, and W. H. McDowell. 2023. Forest growth responds more to air pollution than soil acidification. PLOS One **18**:e0256976.)

This comment is also applicable to p. 4-13; although the studies cited describe the effects of deposition-related changes in soil chemistry on tree growth, direct effects of oxides of sulfur and nitrogen can also be important in affecting tree growth.

Page 4-24: While there is good evidence of changes in terrestrial ecosystems with N addition experiments, long-term patterns in N availability suggest that N availability is declining in relation to N demand in New England. Mason, R. E., et al. 2022. Evidence, causes, and consequences of declining nitrogen availability in terrestrial ecosystems. Science **376**:eabh3767.

Chapter 5 – Exposure Conditions Associated with Effects

Does the Panel find the presentation of information and quantitative analyses related to welfare effects of oxides of nitrogen, oxides of sulfur and particulate matter to be technically sound and clearly communicated? What are the Panel's views on the quantitative analysis of aquatic acidification risks, including the choice of analysis approach and spatial resolutions? Does the document appropriately characterize the results of the analysis, including key limitations and associated uncertainties?

The chapter's information and quantitative analyses related to welfare effects of oxides of nitrogen, oxides of sulfur and particulate matter are in general technically sound and clearly communicated. The choice of quantitative analyses to address aquatic acidification risks and its spatial resolution is appropriate. The key limitations and associated uncertainties are discussed in sufficient detail.

Specific comments:

Ch. 5 5-3 line 4: “intended to reach beyond individual critical loads developed over a variety of studies and ecosystems and consider the underlying study findings with regard to key aspects of the environmental conditions and ecological characteristics studied.” This is an awkward statement, perhaps edit to “intended to reach beyond individual critical loads developed over a variety of studies and ecosystems and instead to develop a broader understanding of the environmental conditions and ecological characteristics that result in variation of critical loads among ecosystems.”

Page 5-3 line 32 “These studies, however, do not generally report the SO₂ concentrations in ambient air or account for the influence of changes in concentrations of co-occurring pollutants such as ozone” but see Hruska et al. for a study that addresses some aspects of this. Hruška, J. et al. 2023. Forest growth responds more to air pollution than soil acidification. PLOS One **18**:e0256976.

Page 5-10 line 5; “is relatively pH sensitive” should be “is relatively pH insensitive” based on the quote and figure

Page 5-1- line 26 is unclear; is the “high S absorption” what remains (sorption capacity) or is it the high levels of S that remain adsorbed on the soils? The example in line 27 does not necessarily illustrate either interpretation, as it speaks to hydrologic routing that can bypass soils almost entirely.

Page 5-12 and throughout: italicize species names e.g. *Salvelinus fontinalis*

Page 5-18 lines 12, 14: NO_3^- should be NO_3^-

Page 5-19 line 6 hectare usually abbreviated as ha, not Ha

Page 5-44 line 18; model runs to 2011 (the past) or 3000 (900 years from now) seem like an odd choice. Verify the dates are correct

Page 5-69 line 14 BC:AL should be BC:Al

In general, citation of any recent papers that link changes in deposition of NH_4^+ to PM 2.5 levels and trends, and papers showing changes in deposition associated with climate change would be very helpful. For example, a recent paper shows striking changes in the proportion of oxidized vs reduced in total N wet deposition associated with snow/no snow events, and increases in dissolved organic N concentrations over time in precipitation (Murray et al. 2022; Nitrogen wet deposition stoichiometry: the role of organic nitrogen, seasonality, and snow. *Biogeochemistry* 2022 Vol. 160 301-314 DOI: 10.1007/s10533-022-00966-00). These changes may be related to levels of precursor PM 2.5. Although not addressed in this specific paper, the links between changes in deposition of reduced N that are likely associated with PM 2.5 dynamics should be highlighted as an area of uncertainty and a need for future research to protect the public welfare.

Chapter 6 – Relationships of Deposition to Air Quality Metrics

To what extent does the Panel find the investigation of relationships between air concentration and deposition to be technically sound and clearly communicated? What are the CASAC views on the consideration of key limitations and associated uncertainties?

The approach used in this chapter is clearly presented. Overall, however, the approach seems to be more of a scattershot analysis or fishing expedition, rather than testing specific hypotheses about relationships among various measurements, and measurement approaches, that then might be used to inform policy.

For example, Figure 6-11. Why is everything plotted against everything else? Is there no a priori expectation or no fundamental physical attributes of the atmospheric system that would lead to specific predictions? I would find it more compelling if more emphasis was placed on documenting and understanding the strongest relationships, rather than simply portraying them all.

A key limitation of the current presentation is that it focuses largely on the relationships between drivers and responses during the entire period of record. This is quite valuable to understand how the current situation has evolved over time and the fundamental drivers that have shaped the welfare effects at the

decadal scale. In addition to this important retrospective analysis, however, it is critically important to focus on the current situation. Changes in the predictability of drivers and responses have occurred as overall N and S deposition has changed. The relationships among measured variables are often different in the most recent years, making it unclear whether past decadal-scale drivers or proxies will be useful in the coming decades.

More specific comments:

Figure 6-17. Are the linear fits all statistically significant? They sure don't seem to be. Remove the trend lines for those that are not statistically significant.

Figures 6-23 and following figures until 6-28. What gives the extraordinarily odd distributions of the data in 6-23 and 6-24? Shouldn't that be addressed in more detail? Why are the figures not consistent, what is meant by using weighted average versus average in 6-25 and 6-26? I can make a guess, but it should be made clear.

Dr. Hans Paerl

Comments and References for Chapters 4 & 5

I am commenting specifically on Chapters 4 and 5, which overlap and could potentially be combined in one chapter. Overall, greater emphasis is needed with regard to the importance of atmospheric deposition of nitrogen (ADN) as a “new” source N entering the freshwater to marine “continuum”. Estimates range from ~20 to over 40% of total N loads, when we consider both indirect (i.e. entering via the watershed) and direct deposition onto the water body (Jaworski et al. 1997; Paerl et al. 2002; Whittall et al. 2003). Studies have shown ADN to be an important N source supporting both freshwater and marine eutrophication (Paerl 1995, 2002; Paerl et al. 2002; Burns et al. 2021), and associated water quality degradation, including harmful algal blooms (HABs), bottom water oxygen depletion, fish and shellfish kills (Paerl and Barnard 2020). With regard to ADN composition, NO_x is a major source of biologically available N, but both organic N and NH_x are increasing relative to NO_x, especially in agricultural, urban and even forested watersheds (c.f., Paerl et al. 2020). Therefore, with regard to human welfare effects (safe potable, fishable and recreational water supplies), ADN plays an integral, and in some regions (N limited estuarine and coastal waters downwind and downstream of ADN inputs), increasingly-important role.

While nitrogen limitation and hence sensitivity to N enrichment characterizes many of our estuarine and coastal waters (Nixon 1995), recent work has shown that due to the combined impact of legacy phosphorus loading and retention along the freshwater to marine continuum, there has been a noticeable shift in the nutrient limitation paradigm in the freshwater component, from exclusive phosphorus limitation of primary production in freshwater systems (c.f. Schindler et al 2008) to either N and P co-limitation or even N limitation (Elser et al. 2007; Howarth and Paerl 2008; Conley et al., 2009; Paerl 2009; Lewis et al. 2011; Paerl et al. 2016, Wurtsbaugh et al. 2019). Examples include some of the most iconic large lake systems experiencing eutrophication and HAB problems, including Lakes Erie (USA-Canada), Okeechobee (FL), Utah Lake (UT), Great Salt Lake (UT), the NY finger lakes, and large reservoir systems (e.g., Jordan and Falls Lakes, NC, numerous midwestern and western lakes and reservoirs) as well as freshwater components of the San Francisco Bay and Mississippi/Atchafalaya Delta system (LA). These systems serve as major sources of drinking and irrigation water, and provide fishing and recreation resources for millions of residents and visitors.

With regard to expanding HAB (and specifically toxic cyanobacterial) outbreaks in these systems, both NO_x and NH_x represent major sources of N supporting the production of N-rich cyanotoxins, including microcystins, cylindrospermopsin and nodularin, encountered in water supplies (Davis et al. 2015; Gobler et al. 2016; Chaffin et al. 2018; 2020). In particular, NH_x is a potent stimulant of HAB biomass and toxin production (Newell et al. 2019), and hence warrants attention with regard to its role as an ADN source.

Overall, atmospheric reduced N emissions/deposition are increasingly important from both budgetary (loads), wildlife and human welfare effects. *Will increasing wild-fires (due to climate change impacts) affect this as well?*

Also, aquatic acidification in response to atmospheric N enrichment leading to eutrophication needs to be reassessed. In some cases where highly eutrophic conditions and algal blooms are stimulated, "basification" can also result (not just acidification) (Baumann and Smith 2017; Hall et al. 2023-in

revision), and this requires further examination and clarification with respect to biogeochemical, ecological and human resource (water use, fisheries) perspectives.

Lastly, the relative importance of ADN as a “new” N source can increase downstream of the axis of estuaries and off-shore of coastal sounds and bays, where a bulk of the terrestrially supplied N has been assimilated by primary producers (Paerl 2002, 2009). This needs to be incorporated in loading estimates, spatial modeling of ADN inputs, and linkage to eutrophication and HAB potentials (Paerl and Piehler 2008) (Fig. 1).

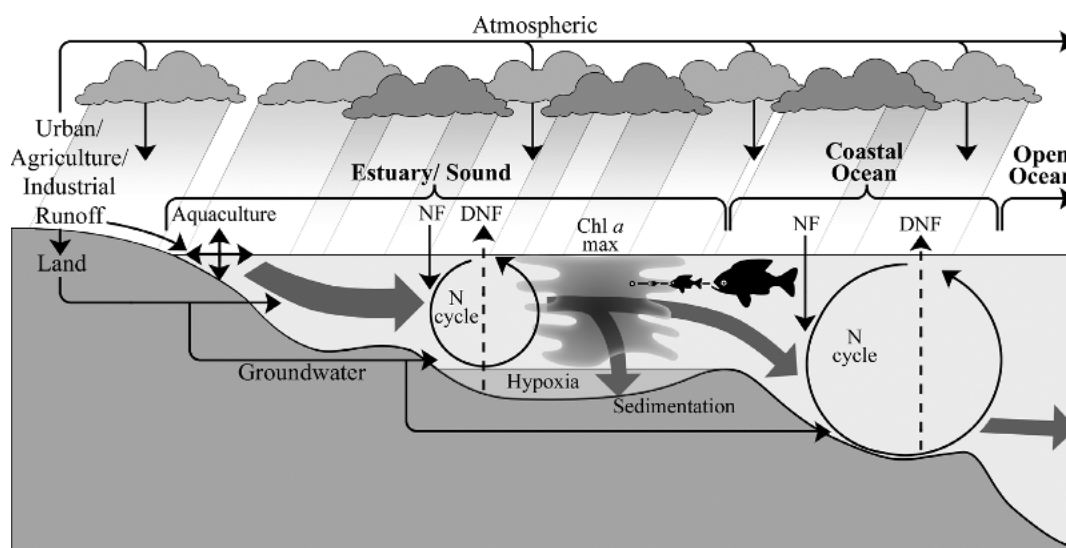


Figure 1: Schematic diagram illustrating sources, transformations and fates of nitrogen (N) along the estuary to ocean continuum. Surface, subsurface and atmospheric pathways of externally supplied or new N inputs attributable to anthropogenic activities are shown as are internal N cycling. The combined anthropogenic N inputs are shown as a thick arrow (upstream), which decreases in thickness downstream as a portion of the N inputs settles to the bottom sediments and is buried and/or denitrified (DNF). NF represents nitrogen (N₂) fixation, a biologically-mediated new N input. The linkage of anthropogenically-enhanced N inputs to accelerated primary production or eutrophication, and its trophic and biogeochemical fate are also shown. In many estuarine and sound systems, primary production and phytoplankton biomass are maximal in mid-system locations, where adequate new N loads and decreasing rates of flushing (increasing residence times) overlap. The resultant chlorophyll a maximum (Chl a max) is characteristic of estuarine systems. This zone can be bypassed by atmospheric inputs. Figure is from Paerl and Piehler (2008).

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Mr. Richard L. Poirot

Chapter 2 –Air Quality

To what extent does the Panel find that the information in Chapter 2 is clearly presented and that it provides useful context for the review?

The information on air quality & deposition in Chapter 2 is clearly presented in sufficient detail to provide useful context for this review. Several of the figures could be clearer and some new figures and/or explanation could be helpful. For example, Figure 2-39 presentation of average 2019-21 S deposition employs a scale of 0-20 kg/ha to show spatial deposition patterns which are generally less than 5 kg/ha everywhere. Variations in these lower, recent deposition rates are most relevant to the recent exceedances of critical S loads presented in Chapter 5. The Figure 2-26 PM_{2.5} composition pie chart showing spatial differences in recent PM_{2.5} composition is not clearly explained. I assume, but don't know, that the "sulfates" and "nitrates" slices include associated ammonium, and that the "OC" slice represents "organic matter" including estimated O and H associated with OC? Since you later suggest PM_{2.5} mass or selected components as possible indicators, it would also be useful to convey how the relative magnitudes of these PM_{2.5} components vary spatially, seasonally and over time.

For example, **Figure 1** below shows average 2001 PM_{2.5} mass composition at IMPROVE Class I and IMPROVE Protocol sites (excluding several urban sites collocated with EPA CSN). It's analogous to Figure 2-26 in the PA, except the pies are sized relative to total reconstructed PM_{2.5} mass and based on 2001 data. Twenty+ years ago, ammonium sulfate and ammonium nitrate were predominant components of PM_{2.5}, especially in the East where they accounted for 1/2 to 2/3 of PM_{2.5}, making PM_{2.5} mass a strong indicator of 2001 S and N deposition. **Figure 2** below is similar but based on 2021 data. Ammoniated sulfates and nitrates accounted for a quarter to half of PM_{2.5} in the East and smaller fractions in the West, as organic matter is becoming the predominant PM_{2.5} species. Changes in PM_{2.5} mass have become less indicative of changes in aerosol S and N compounds than they used to be, and this trend will continue in the future as further reductions in S and (oxidized) N emissions occur.

Figure 3 below is also based on 2021 IMPROVE data but reflects fractional PM_{2.5} species contributions to PM_{2.5} light extinction. Fractional contributions of hygroscopic S and N compounds to PM_{2.5} light extinction are proportionately greater than their fractional contributions to PM_{2.5} mass, making this a **better S and N indicator** than PM_{2.5} mass. PM_{2.5} light extinction was advocated by EPA staff and strongly supported by CASAC as the preferred indicator for a short-term (sub daily) secondary PM_{2.5} standard to protect visibility (in non-Class I areas) in the 2012 PM NAAQS review. It would also be a better long-term indicator for ecological effects than PM_{2.5} mass. **Figure 4** shows even greater fractional contributions of S and N compounds of PM (including coarse PM) light extinction on the 20% of the "most impaired" days in 2021. The most impaired days are screened to exclude days with episodic "natural" forest fires and dust storms; and gradual improvements to this subset of days is an important basis of EPA's Regional Haze Regulations for Class I areas. This group of days would also be a **much better indicator** than average PM_{2.5} mass for ecological effects of S and N Deposition.

Figure 1. Average 2001 PM_{2.5} Mass at IMPROVE and IMPROVE Protocol Sites

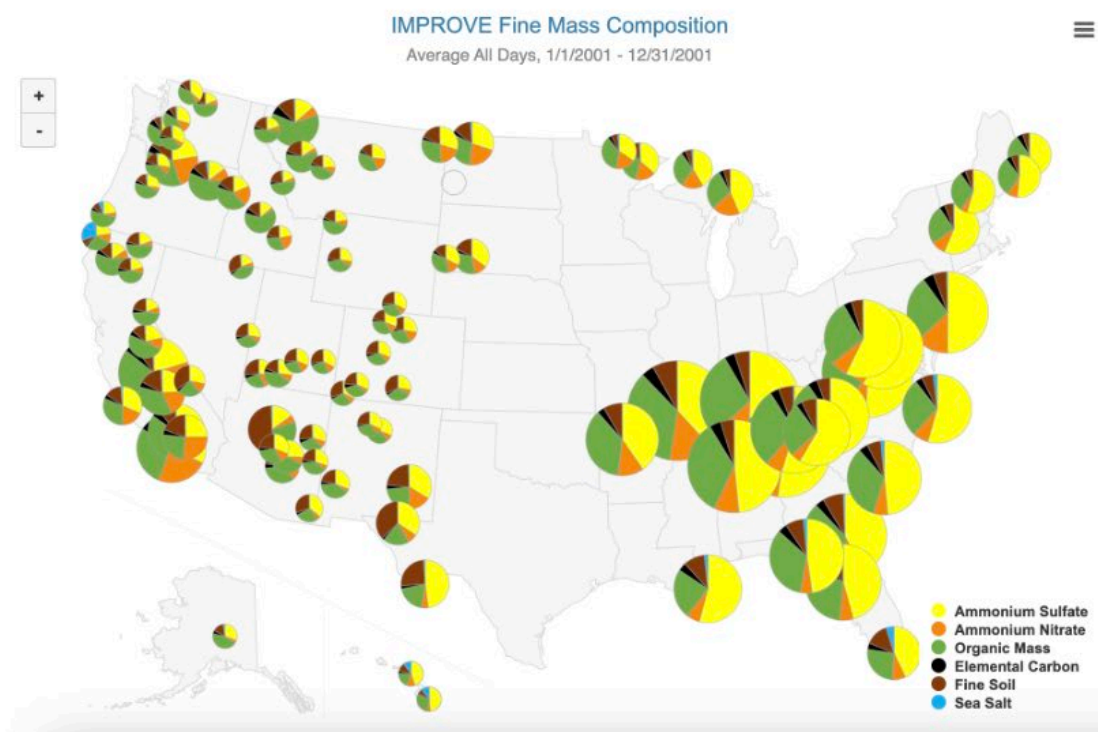


Figure 2. Average 2021 PM_{2.5} Mass at IMPROVE and IMPROVE Protocol Sites



Figure 3. Average 2021 PM_{2.5} Light Extinction at IMPROVE and IMPROVE Protocol Sites

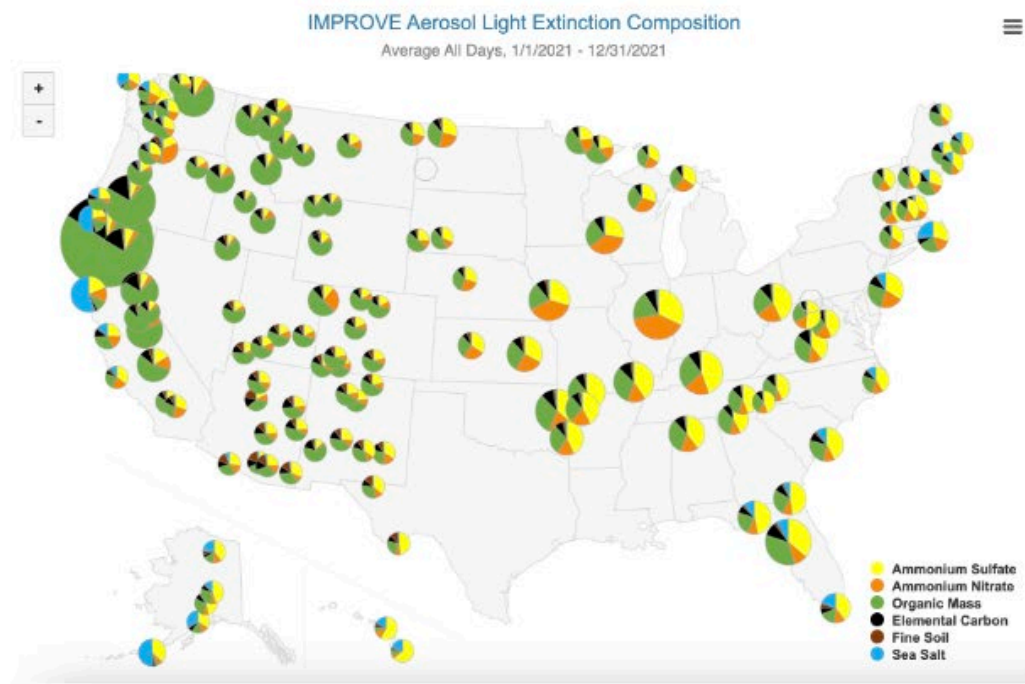
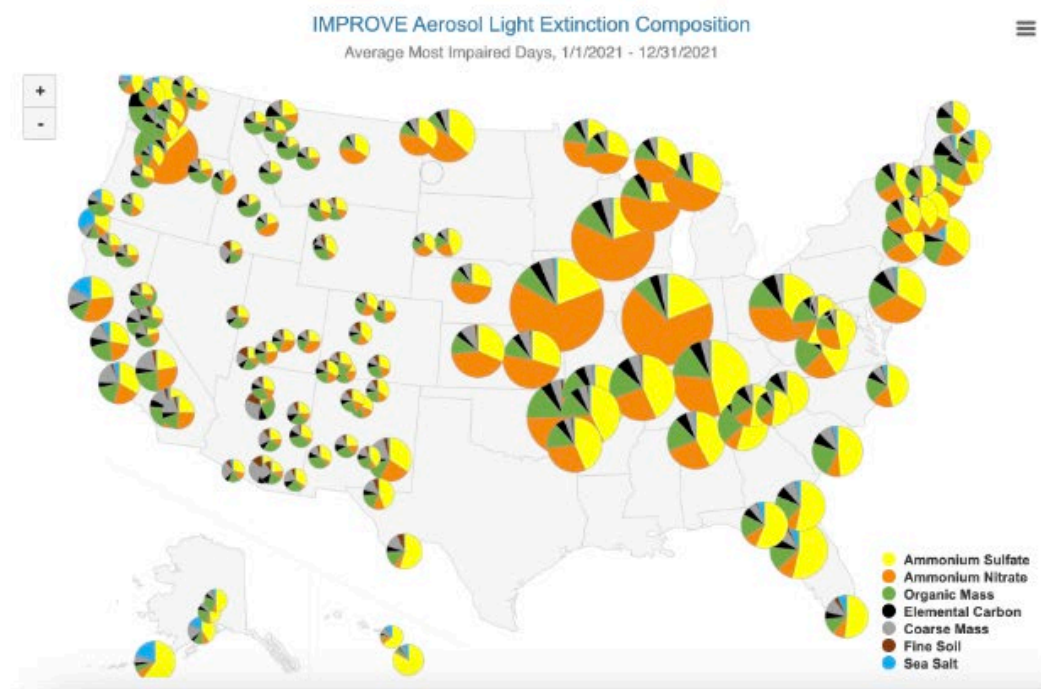


Figure 4. Average 2021 PM Light Extinction on 20% Most Impaired Days (includes Coarse PM)



Figures 1-4 from: <https://views.cira.colostate.edu/fed/Express/AqrvTools.aspx#Visibility>

If PM_{2.5} mass and/or PM_{2.5} effects (like light extinction) or routinely measured PM components are being considered as indicators for a PM-related indicator which best reflects S and N deposition, The best, most relevant indicators might be PM_{2.5} S (directly measured in both EPA CSN and IMPROVE networks), and PM_{2.5} N, including N from NO₃ as well as N from NH₄, associated with NH₄NO₃ and (NH₄)₂SO₄ (measured and/or estimated by comparable methods by both IMPROVE and CSN networks).

Other graphics that could be useful in this chapter include a time series of 3-hour SO₂ design values – since that is the current secondary standard. A scatter plot of 1-hour vs. 3-hour SO₂ design values for recent years might help show how ineffectual (weaker than the primary) the current secondary standard is. Scatter plots of hourly vs. annual NO₂ and SO₂ might also reveal useful features of the data distributions and provide a link to averaging times most relevant to deposition. The observation that SO₂ and NO₂ levels have continued to decline even at levels well below the secondary NAAQS shows that other considerations (primarily attaining lower PM_{2.5} levels) have been responsible for past reductions in S and N concentrations and deposition, and helps support your consideration in later chapters that PM_{2.5} components might be good future indicators for S & N deposition-related standards.

Aspects of reduced N spatial and temporal concentrations and deposition patterns could be presented in more detail. You could show a national concentration maps and trends in PM_{2.5} NH₄ and total PM_{2.5} N either based on estimates from IMPROVE and CSN SO₄ & NO₃ and NH₄ or from CSN measurements (or both). This will show steep declines over time even as NH₃ emissions have increased. The TDEP measure/model estimates show substantial increases in total NHx deposition over the past 20 years. How does this compare to the estimated emissions trend? It appears this deposition increase has been entirely due to increases in wet deposition, as there's been almost no change in estimated dry NHx deposition over the past 20 years. Some added discussion and explanation of these patterns would be useful. Additional comments on Chapter 2 are in my line by line comments below.

Chapter 7 – Review of the Standards

What are the Panel views on the discussion of key considerations in sections 7.1 and 7.2 and preliminary conclusions? What are the CASAC views regarding the areas for additional research identified in Chapters 3 and 4? Are there additional areas that should be highlighted?

The section 7.1 summary of the relative absence of current ecological effects from direct exposures to SO₂, NO₂ and PM is reasonable. The observation that there hasn't been an exceedance of the secondary NO₂ standard for the past 30 years (while emissions, concentrations and deposition continued to decline) illustrates that recent benefits from NOx decreases have been unrelated to the current NO₂ secondary standard. It could be helpful to include a similar statement regarding the attainment of the secondary SO₂ standard.

The section 7.2 summary of relative effects from deposition of S and N also seems reasonable, although I defer to others on the panel with much greater expertise in the various effects areas. For acidic deposition on aquatic ecosystems, the decision to focus the quantitative assessment on S-only appears to be justified by the relatively few waterbodies identified in Appendix 5A for which N deposition contributed to recent CL exceedances. Although it also appears that a similarly small fraction of (a larger number of) waterbodies had influential N contributions 20 years ago. Does this call into question the logic of combining secondary NAAQS reviews for ecological effects of SOx and NOx in the future. It

might make more sense to consider combined effects of these 2 pollutants over a broader range of welfare effects, including visibility.

The extensive analysis of waterbody CL exceedances at different ANC thresholds in different ecoregions and over different recent and past time periods is impressive, though a bit hard to follow in a way that helps support a specific conclusion. The analysis does illustrate a beneficial chemical shift across a wide range of waterbodies with variable historical conditions. Given the extensive historical chemical records of thousands of waterbodies with associated changes in deposition, it might have been informative to include a specific focus on waterbodies that once exceeded and now no longer exceed CL thresholds. Show us the changes in lake chemical composition that resulted from changes in deposition. Perhaps this kind of analysis could help reduce current uncertainties about variable rates of soil weathering that also seem to complicate assessments of terrestrial acidification effects.

The summarization of effects from N deposition is more qualitative than for S. Acidifying N deposition adds to the effects of S in many impacted waterbodies, regardless of whether any specific ANC threshold is exceeded. Nitrogen enrichment effects on aquatic and terrestrial ecosystems are extensive but more complex and more difficult to quantify on a national basis. While some terrestrial plant species may experience growth “benefits” from N enrichment, such benefits come at the expense of other less nitrophilic species in natural ecosystems that are not monocultures. It was disappointing to see no attempt at quantitative assessments of N deposition effects (especially compared to the highly detailed quantitative assessment of S deposition effects for many various thresholds and time periods). The recent Pavlovic et al. (2023)²⁹ analysis described in Dr. Driscoll’s comments should be very helpful in this regard. The National Park Service comments also summarize and provide access to details of N deposition and N CL exceedances for multiple ecological endpoints and for multiple thresholds, time periods, and rural/remote receptors, including many/most “Class I Federal Areas”. Since you have emphasized the importance of protecting air quality-related values in these Class I areas elsewhere in this PA document, the NPS assessments seem especially relevant.

The chapter discusses a range of potential indicators that would link S and/or N air quality to S and/or N deposition and which might be considered to reduce the ecological impacts of these pollutants, or at least help assure that current effects don’t worsen. For S, the current 3-hour SO₂ standard is attained everywhere and therefore provides no protection. Lowering it could have some beneficial effect, but the 3-hour focus on relatively extreme values is the wrong averaging time and may focus on the wrong (few) sources. An annual averaging time is much more logical. As shown in chapter 2, only a few SO₂ monitors currently exhibit annual averages greater than 10 ppb and an annual average set at this level would only influence those 2 monitors (which may also exceed the more time-constrained primary NAAQS) and would help assure that current S deposition rates remain below 5 kg/ha. The current annual secondary standard for NO₂ also provides no protection, as it has been attained everywhere for about the past 30 years. It could be reduced to about 30 ppb without causing any new non-attainment and would help assure that deposition of oxidized N would not increase in the future.

Other potential indicators include PM_{2.5} mass, which includes both particulate sulfate and nitrate, as well as the reduced N (NH₄) associated with these pollutants in PM_{2.5}. It also includes other species, most notably particulate organic matter which is likely to compose an increasing fraction of PM_{2.5} mass in the future. More focused PM indicators might include PM_{2.5} sulfate, nitrate, and associated ammonium. PM N (or PM S+N) would include the NH₄ associated with both SO₄ and NO₃ in PM_{2.5}. “Total” gaseous and PM-phase S and/or N, such as is measured at CASTNET sites (and as an indicator proposed in the previous secondary NO_x/SO_x review) are other potential indicators. All of the above have strengths and

weaknesses, although it could be noted that all of the above have decreased over the past 30 years in parallel with (and resulting in decreases in) S and N deposition.

While the CASTNET measurements show (and should show) slightly better correlations with S and N deposition than other gaseous or PM indicators, CASTNET also has the fewest sites, has lost more sites recently and would require considerable effort to develop their standard procedures into federal reference methods. The gaseous SO₂ and NO₂ indicators have the advantage of existing FRM networks, but the disadvantage of missing reduced N, which contributes a large and growing fraction of N deposition. PM_{2.5} mass also has the advantages of a large existing FRM network and includes particle-phase versions of all the relevant S and N species. A disadvantage is that it also includes a large and proportionally increasing fraction of organic matter.

Speciated PM_{2.5} measurements are routinely available from a large number of sites in the CSN and IMPROVE networks. The relevant PM species (SO₄, NO₃ and NH₄ – or PM_{2.5} S and N) would be much more direct indicators of S & N deposition than total PM_{2.5} mass. There would be issues combining the State-operated and urban/suburban CSN with the largely FLM-operated rural/remote IMPROVE (although that would assure measurements most relevant to the sensitive, protected Class I areas). The IMPROVE data are also used by states in complying with the EPA Regional Haze Rule, and there have been extensive and continuing efforts to compare and harmonize methods between these networks. It should also be noted that EPA Staff had recommended and CASAC had supported a “reconstructed PM light extinction” metric utilizing the speciated PM_{2.5} data from the CSN network as an indicator for a visibility-based secondary PM NAAQS in the 2011 PM Policy Assessment.³⁰ So there should be no problem using various combinations of PM species. We could call it something like “acidifying PM_{2.5}”, expressed as the sum of PM_{2.5} sulfate, nitrate and ammonium, or S+N. A PM_{2.5} light extinction indicator (especially one focused on the most impaired days) would also relate more strongly to S & N deposition than PM_{2.5} mass does, since it emphasizes the influence of the hygroscopic S and N species relative to OC and crustal components. We could have an annual standard for deposition effects and a short-term standard for visibility...

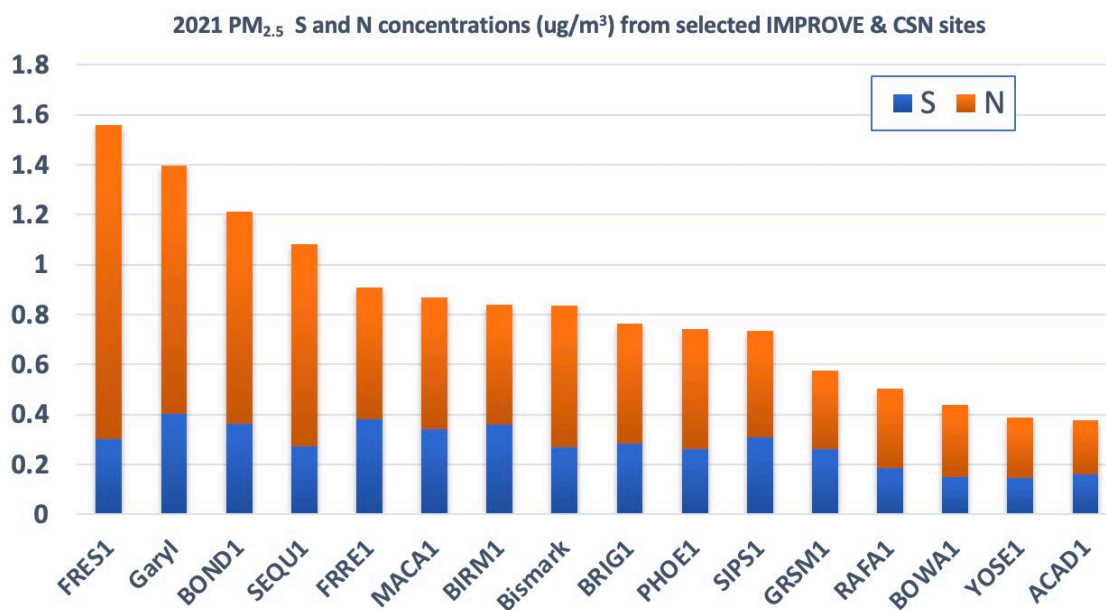
The preliminary conclusions presented in section 7.3 indicate that while there is no clear evidence of direct foliar effects at concentrations below the level of the current secondary SO₂ or NO₂ standard, the analysis of acidifying effects and N enrichment effects of S and N deposition calls into question the adequacy of the current secondary SO₂ and NO₂ standard. A range of preliminary policy options are presented, including retaining the existing secondary standards for SO₂, NO₂ and PM_{2.5} (i.e. do nothing). Ranges of alternative secondary SO₂ standards are proposed, including 200-400 ppb for 3 hours and 10 to 23 ppb as an annual average. Even at the bottom of these ranges, it looks like there would only be 4 sites exceeding 200 ppb for 3 hours and only 2 sites exceeding an annual average of 10 ppb. It's not clear if (but seems likely that) those sites also exceed the primary NAAQS, then exceeding the (more lenient) secondary NAAQS would not impose a burden on those sources. However, secondary standards at the bottom end of the suggested ranges could be a useful mechanism to prevent backsliding. An option for lowering the current secondary NO₂ standard from 53 to as low as 40 ppb is also proposed. The lower end of this range has not been exceeded at any site since 2002; 35 hasn't been exceeded since 2005 and 30 has only been exceeded at a few sites in recent years. A revised annual NO₂ NAAQS at or below the bottom of the recommended range would not achieve much in terms of emissions controls but could help prevent backsliding toward more injurious former conditions.

An alternative secondary PM_{2.5} mass standard in the range of 15 to 12 is also suggested. While a number of sites currently exceed both 15 (level of the current secondary standard and 12 (level of the current

primary), lowering the NAAQS to 12 imposes no new burdens since those sites in the 12-15 range already exceed the primary. For the same reason, a secondary of 12 isn't likely to result in any further reductions in S or N emissions or deposition. It would however correct what many consider to be a poorly justified 2012 decision to set the secondary at a higher, (and therefore ineffectual) level than the primary. Lowering the secondary to 12 would be consistent with the analysis of ecological effects and consideration of appropriate indicators – including those that capture influence of reduced N – reviewed in this document. It would also be more protective of other PM welfare effects, including visibility, which was not effectively addressed in the 2012 and subsequent NAAQS reviews of the secondary PM_{2.5} NAAQS. Depending on the timing of this NAAQS review and pending a final decision on the (primary) PM_{2.5} NAAQS (possibly to 9 or 10 ug/m³), consideration should also be given to lowering the secondary NAAQS to that same level. Otherwise, it will once again serve no useful purpose.

In my opinion, PM_{2.5} S and N (or S+N) from the combined IMPROVE and CSN networks would be the best air concentration indicator(s) to represent S and N deposition using data that's readily available from existing widespread networks (>300 sites, comparable to the number of SLAMS monitors for SO₂ and for NO₂). A very quick look at the data suggests that a PM_{2.5} S+N range of about 1 +/- 0.25 ug/m³ would be worth exploring.

The following figure is based on rough calculations from 2021 data extracted from FED for selected IMPROVE and (a few) STN sites – including several – urban and rural I expected to be relatively polluted (Fresno, CA; Gary, IN; Bondville, IL; Frostburg Reservoir, MD; Birmingham, AL; Sequoia NP, CA; etc.).



Although there are exceptions, the highest S sites are not typically the highest N sites. A combined PM_{2.5} S+N indicator could direct focus to areas where one or the other or both pollutants are problematic, and would also place relatively greater emphasis on the greater continuing problems associated with N deposition. I'm guessing relatively few areas of the country would exceed a combined S+N threshold of about 0.75 or 0.8 ug/m³, and those that did would be in areas where decreased N and/or S emissions would have beneficial effects on S and N deposition on sensitive downwind receptors.

An alternative approach might be to combine a new, restrictive annual SO₂ standard with a PM_{2.5} N (or PM_{2.5} NH₄) standard to apply maximum pressure on currently unregulated NH_x emissions. The previously mentioned PM_{2.5} light extinction indicators are also worth a closer look.

Deposition-Based Standards

In the previous secondary SO_x/NO_x NAAQS review, EPA had considered – and rejected – the idea of using a deposition-based indicator for a secondary standard, the general argument being that the NAAQS must relate to the presence of the pollutants in the ambient air. I think this argument could be reconsidered. With the exceptions of effects on climate and visibility, virtually NO health or welfare effects of ANY criteria pollutants result directly from their “presence in the ambient air”. Nearly ALL effects result from Deposition of the pollutants – within the human respiratory tract, within plant tissues, on surfaces followed by hand-to mouth ingestion for Pb, and on plants, soils, surfaces, waterbodies, etc. for N, S and PM.

Deposition is also a logical and direct indicator of the presence of the pollutants in the ambient air (but-for causation), and is arguably a superior indicator of the presence of pollutants in the “relevant column” of the ambient air, compared to a point measurement from a single breathing height air monitor. Wet deposition reflects the pollutants presence throughout a much deeper column of the ambient air, most relevant to welfare effects. Although there’s no definition (that I could find) of “ambient air” in the Clean Air Act, EPA regulations (40 C.F.R. § 50.1(e)) define “ambient air” to mean “that portion of the atmosphere, external to buildings, to which the general public has access.” This definition has been slightly modified on several occasions through EPA guidance, most recently in 2019 to further exclude air over private property to which public access is restricted (Wheeler, A., 12/2/2019, Memo to Regional Admins. on Revised Policy on Exclusions to Ambient Air).

“Air to which the public has access” is an appropriate concept for consideration of human exposures and associated health effects. However, it’s poorly suited for considering welfare effects, few of which are dependent on whether the public has direct access to the air. For example, PM effects on climate, visibility; ozone effects on UV radiation or on crops or trees; and various effects of S and N that result from atmospheric deposition. Wet deposition of S and N aggregates pollution present in clouds and the entire column of ambient air beneath them. It would be useful for EPA to consider modifications to this definition to better reflect the nature of welfare effects.

For example: “For purposes of evaluating pollutant effects on public welfare, ambient air also includes any portion of the atmosphere in which pollutants are present that may reasonably be anticipated to cause or contribute to effects on public welfare, such as through modifications to incident solar radiation or through deposition to the environment”. If the ambient air can include portions of the atmosphere above just “what the public has access to”, then deposition could clearly be considered a better indicator of the pollutants present in that column of than a point air quality measurement from a near-surface monitor – and would also better reflect the pollutant effects on public welfare.

A total (wet + dry) deposition-based standard that includes reduced N would be appropriate since it would capture both an important precursor (NH₃) and important component (NH₄) of PM. This will continue to be true, even as the NO₃ and SO₄ (and NH₄) components of PM continue to decline in the future. However, while an indicator like PM N will become a progressively weaker indicator of reduced N in the future (as it’s partner anions decrease), total N deposition (including wet) will continue to

strongly reflect the presence of NH_x in the ambient air, providing a better link to this important but otherwise unregulated pollutant.

The current Policy Assessment and the supporting Integrated Science Assessment both depend heavily on annual estimates of total wet and dry deposition of S and N species produced by the NADP Total Deposition Science Committee (TDep). The TDep approach, initially described by Schwede and Lear (2014), combines state of the art monitoring and modeling data to produce the consensus highest quality annual estimates of wet, dry and total deposition of various S and N (and other relevant) species, currently with 4 km spatial resolution over the coterminous US. This approach has been continually refined over the past decade, with improvements carefully described in annual TDep methods documentation files (NADP, 2023).

The measurements, modeling and methods for blending them and assessing their results are conducted, supported and refined by a wide range of scientists from states, academia, EPA, multiple other federal agencies and Environment Canada. Results from a significant recent enhancement of TDep methods and results will be presented shortly for peer review by Beachly et al., in preparation). Given that the objective of a secondary NO_x/SO_x/PM NAAQS indicator would be to represent annual deposition over broad spatial areas (ecoregions), the annual TDep estimates are uniquely and ideally suited for this application.

Traditionally, EPA has relied exclusively on near-surface air quality measurements, with formal “Federal Reference Methods” developed by EPA and delegated to the states, as the bases for determining compliance with primary and secondary NAAQS. Given the clear importance of deposition to ecological S, N and PM effects, and the complexities and uncertainties associated with linking near-surface air quality measurements to regional deposition, the TDep data provide a superior approach for assessing both temporal and spatial deposition of S and N. The continuing multiagency support for NADP TDep calculations helps assure continued future scientific and logistical support for the future.

Research Needs

The stated research needs all look reasonable. Some possible additions:

- Continued research into application of remote sensing techniques to quantify total column SO₂, NO₂, ammonia, PM_{2.5}, and carbonaceous aerosols, and to compare and merge these data with surface observations and air quality models.
- Continued development of measurement methods to quantify gaseous and particulate S and N and other PM_{2.5} species, particularly those that might be applied to large existing monitoring networks.
- Additional analyses of chemical and biological changes that have occurred in sensitive aquatic and terrestrial ecosystems in conjunction with historical changes in S and N deposition.

Line-by-line comments

P. 1-7, lines 1-6: This is awkward wording and misses an important point. The phrase “...based on the original AQCD and recognition of effects on vegetation and to match the primary standards...” sounds like there was some additional recognition of vegetation effects besides what was considered in the 1969

CD and also makes it sound like the secondary standards were set to match the primary – neither of which is true. What about rephrasing to something like:

“The EPA first established primary and secondary NAAQS for PM in 1971 (36 FR 8186, April 30, 1971), based on the original AQCD (U.S. DHEW, 1969b). The secondary standards were set at 150 $\mu\text{g}/\text{m}^3$, 24-hour average, for total suspended particles (TSP), not to be exceeded more than once per year, and 60 $\mu\text{g}/\text{m}^3$, annual geometric mean. *[although see following comment on whether the 1971 annual mean secondary was actually a “standard”]*. These secondary standards were more stringent than the primary standards and based on consideration of PM effects on visibility, materials and vegetation, as summarized in the 1969 AQCD.”

P. 1-7, lines 7-8: Here's a minor point, but there's something slightly squirrely about the 1971 annual TSP secondary NAAQS: its sometimes mentioned and sometimes not. It's not mentioned in John Bachmann's classic (and otherwise thoroughly detailed) 2007 history of the US NAAQS¹, or in the 2005 EPA PM NAAQS Staff Paper², or in the Federal Register notice announcing 2013 Revisions to the PM NAAQS³ (which include all other past primary & secondary PM NAAQS). It was mentioned (as 60 $\mu\text{g}/\text{m}^3$ annual geometric mean) in many other places, including the original 1971 Federal Register notice announcing the NAAQS, **but** is described there “as a **guide** to be used in assessing implementation plans to attain the 24-hour standard.”⁴ So was it a standard or a “guide”? If it was more of a “guide”, maybe this concept could be useful in the future to establish a kind of somewhat “softer” secondary standard – a kind of guide or long-term goal that wouldn't carry the stigma of secondary non-attainment.

P 1-7, line 15: Since you are emphasizing linkages in causes and effects of SO_x, NO_x and PM, it might be useful to add a few lines here, like:

While the 1987 secondary PM standards were ultimately set equal to the primary PM₁₀ standards, it can be noted that the 1982 EPA PM staff paper recommended consideration of a separate, more stringent secondary standard based on a PM_{2.5} indicator.⁵ CASAC comments were supportive and noted that the strongest case for a secondary PM_{2.5} standard would be “one that links emissions of nitrogen oxides and sulfur oxides with the interrelated aspects of acidic deposition, possible climatological effects, and visibility.” CASAC further advised that “it may be more efficient to consider a common standard linked to fine particles than to establish a separate set of controls for each of these problems and pollutants.”⁶ This was 10 years before a primary PM_{2.5} standard was established.

P. 1-10, lines 1-2: The subsequent PM NAAQS review, completed in 2012, was only partially “responsive” to this court remand. Yes, the annual secondary PM_{2.5} standard was (finally) set at a different level from the primary, but no, a secondary standard set at a level weaker than the primary doesn't protect visibility or anything else. Nor were the indicator, level, averaging-time or form at all consistent with what was discussed in the 2009 PM ISA⁷, the 2010 Urban-Focused Visibility Assessment⁸, the 2021 Final PM Policy Assessment,⁹ or the 2012 PM NAAQS Proposed Rule.¹⁰ Just sort of pulled out of thin air at the last minute...

P. 1-12, line 24: “profound uncertainties”. Really? A word count for “uncertain” (includes ...ty, ...ties) shows 400 instances in this document. I guess that's pretty profound, and certainly gives the Administrator plenty of wiggle room...

P. 2-2, line 15: “for” should be “form”.

P. 2-2, line 21: Can you give any indication of what you mean by “very fast rates” of sulfate production? “...as high as x%/hr...”

P. 2-4, lines 18-19: I don’t think “whether or not a pollutant deposits to plant stomata” is a correct differentiation between wet and dry deposition. There’s plenty of dry dep that doesn’t occur through stomata (to leaf cuticles, bark) or involve plants at all (soils, snowpack, surface waters, etc.).

P. 2-5, line 9: Somewhere in this section (2.2.1) could you mention the form(s) of NO_x emissions. Presumably most of these emissions are from NO. Do you combine NO and NO₂ in these NO_x emissions? What are the units?

P. 2-5, lines 20-21: Are emissions from lightening included in the “other” category of Figure 2-2? If so, 1% seems kind of small. See for example Kang and Pickering (2018)¹¹, Arndt et al. (2019)¹². Are forest fires a component of “biogenics”?

P. 2-7, line 1: You could add “anthropogenic” after “Total”.

P. 2-8, lines 2-15: Should there be a mention of the relatively small fraction of SO_x emissions which are in the form of “primary sulfates” emitted directly from combustion sources? Could be in the range of at least a few % of total SO_x emissions, and possibly more for certain source types See for example Homolya and Lambert (1981)¹³, Holt et al. (1982)¹⁴, Dominguez et al. (2008).¹⁵ As I recall, this used to be kind of a controversial subject (hard thing to measure). Did it get resolved?

P. 2-12, lines 9-10: Do the new emissions models reduce uncertainties or not?

P. 2-20, Fig 2-16: It might be useful to provide an indication of CASTNET sites recently discontinued in 5/22, as well as mention of the current CASTNET network review. I helped provide past advice on reductions to both IMPROVE and STN networks – but I bet in the end you will likely lament the need more monitoring data – because profound uncertainties...

P. 2-21, lines 24-26: For the most part, IMPROVE never routinely measured or reported NH₄. Until about 1999, the nylon filters used for Ionic analysis were extracted using a basic solution of sodium carbonate and bicarbonate to assure complete extraction of aerosol nitrate. This basic extract would have interfered with potential analysis for NH₄. Following an experiment around 2000, IMPROVE started extracting the filters with deionized water (+ filter sonification) - which would have allowed analysis for NH₄, but such analyses weren’t routinely conducted – except possibly for a few sites in the SE US for a few years. I don’t think these were official IMPROVE measurements and the NH₄ data, if any, aren’t reported in the IMPROVE database. I think the rational is the assumption that almost all PM_{2.5} NO₃ is NH₄NO₃, but that NH₄ associated with SO₄ may have formed on the filter during or after sampling if acidic sulfates were exposed to gaseous NH₃. IMPROVE assumes sulfates and nitrates are fully neutralized by ammonium, and this assumed ammonium is included in their light extinction calculations. This has likely become more consistently true over time, as S emissions have decreased and NH₃ increased.

While the CSN network measures ammonium, there are likely to be negative sampling artifacts on the nylon filters used in both IMPROVE and CSN networks (Solomon et al., 2000, Yu et al. 2006).^{32, 33} For the PM_{2.5} light extinction indicator proposed by EPA staff in the 2011 PM NAAQS review Policy

Assessment, EPA proposed using the CSN sulfate and nitrate data, but estimating ammonium using the IMPROVE algorithm ($\text{NH}_4 = 0.375 * \text{SO}_4 = 0.29 \text{ NO}_3$).

P. 2-23, lines 24-28: This is an important point. Conversely, if concentrations were to start increasing again, we might expect effects to worsen. Maybe, if all sites are below NAAQS, a secondary NAAQS could be set at a lower level – to assure no backsliding. I recommend considering this concept for all NAAQS reviews: As measured concentrations generally fall below the NAAQS, the secondary NAAQS should be lowered to prevent backsliding to past levels of more injurious pollutant concentrations. Yes, I know there are “prevention of significant deterioration” requirements in the CAA, but these are a huge pain for states & sources to comply with and get gamed all the time. The recent (2019-21) appearance of high annual average SO_2 levels in Figure 2-25 – exceeding levels that hadn’t been seen in the previous 15 years – is a good example of undesirable backsliding (unless they just resulted from new monitor locations, but still illustrates the concept).

P. 2-25, Figure 2-21: I’m curious at how there appears to be an increase in the highest annual means (Fig 2-21), but not so much in peak 1-hr (Fig 2-22) concentrations, which appears to correspond to the addition of new sites starting around 2014. I’m guessing these are mostly the near-road sites, which don’t really seem to show higher peak hourly levels. If you excluded the near-road sites from comparison to a secondary NAAQS, you could almost cut the annual NO_2 NAAQS in half (to prevent backsliding) and still wouldn’t have any sites out of attainment. Also, as a general consideration, would it be reasonable to consider near-road measurements as relevant to human health effects, but not to ecological effects? Alexandra will likely disagree..

P. 2-27, Figure 2-23: Since there are no violations of the secondary SO_2 NAAQS, it looks like (as with NO_2), you could substantially drop the level of this secondary NAAQS (to prevent a return to the bad old days) and not have any new secondary nonattainment (at sites which don’t also exceed the primary).

P. 2-28, Figure 2-22: Shouldn’t we also see a plot of the 3-hr design value trend – since that’s the secondary standard. It would also be informative to see a scatter plot comparing 1-hr vs. 3-hr as I think (guess) it would clearly show that the current secondary standard is weaker than the primary, and therefor serves no useful purpose.

P. 2-28, Figure 2-25: Are these really “design values”? There’s no annual NAAQS (yet). In the last few years, it looks like only 2 sites had recent averages > 10 ppb. Could you name these sites (assuming they’re the same sites) since they look like outliers? I assume, but don’t know that these 2 sites also exceed the primary, if so (or not), could you indicate by how much, or maybe show their levels as different colored dots in Fig. 2-24. Are these straight annual averages, or 3-year means as you propose as a metric for one of your policy options. Also, it might be informative to show some scatter plots comparing 1-hr and 3-hr peaks vs. annual averages.

P. 2-29, line 5: The level of the current 24-hr primary and secondary $\text{PM}_{2.5}$ NAAQS is 35 ug/m^3 , 98th percentile, averaged over 3 years. The 24-hr PM_{10} primary and secondary standard is 150 ug/m^3 , not to be exceeded more than once per year on average over 3 years.

P. 2-30, lines 18-20: I think trends in several other PM components – like organic matter and elemental carbon - are also distinctly downward at most IMPROVE sites. See for example Jenny Hand’s presentation of the upcoming IMPROVE report from the 2022 IMPROVE Steering Committee meeting

(Hand, 2022).¹⁴ Surely ammonium trends track sulfate and nitrate, and you could show using IMPROVE and CSN NH₄ estimates.

P. 2-29, Figure 2-26: I assume the pie slices labeled sulfates and nitrates also include associated ammonium (If they do, you should say so. If not, these aren't really PM_{2.5} pies. I also assume your "OC" slice represent organic matter, not just measured OC, and also estimates the O & H associated with "particulate organic matter" (POM or 1.6 or 1.8 OC)? Do you also include aerosol water (mostly only present due to SO₄, NO₃, NH₄), and are you trying to describe PM_{2.5} on FRM PM_{2.5} filters (Frank, 2006)¹⁵ or as it exists in the ambient air as quantified in various PM_{2.5} speciation networks?

Generally, I think it's important here to include a clear presentation of your best measurements/estimates of PM_{2.5} ammonium (and other aerosol species like water that would not be there "but/for" sulfates and nitrates). This could also be a good place to show exactly what a few (readily measurable with current networks) S+N PM_{2.5} "indicators" might look like, for example:

- PM_{2.5} sulfate + nitrate + associated ammonium (and maybe water – or not)
- PM_{2.5} mass – smoke – soil – EC – sea salt (how do these 2 compare?)
- PM_{2.5} S, N and S+N.

P. 2-37, lines 4,5: What are "non-air related sources of deposition"? Is this like direct discharges to waterbodies - and is that deposition?

P. 2-40, lines 16, 17: What does "and a small fraction is not included as part of TDEP" refer to?

P. 2-41, lines 22,23: I'm surprised that bias in NH₄ wet deposition is the same as and as high as the bias in NH₃ concentration – since NH₄ wet deposition is more widely measured and less spatially variable. Are you sure you don't mean 55% bias in NH₃ concentration and dry deposition?

P. 2-43, Figure 23-9: There's no reason why depositions of S and N need to be compared with each other on a 1:1 basis (both scales of 0-20), so why not show S with a lower scale that would show some pattern? This presentation seems especially illogical when you subsequently claim that it's only S deposition that contributes to acidification. It also makes it impossible to make sense of any of your subsequently described CL thresholds.

P. 2-45, Figure 2-40: The figure caption refers to the % N deposition as reduced N (gas phase NH₃ and particle phase NH₄⁺). But I assume this likely also includes wet NH₄⁺ deposition, since particle-phase NH₄⁺ doesn't account for much deposition directly – except maybe at higher elevations in the West. Keep the particle phase deposition in there if it was part of the calculation, but I think you need to add wet. Or maybe this shows "% of dry N deposition", although I think % total N dep is a more important concept.

Also, it might be informative to show & contrast trends in particle phase NH₄ – which should basically be the same as trends in the sum of particle SO₄ and NO₃ and strongly downward – with trends in wet NH₄ which I think are generally upward. You can't really see much trend in NH₃ with your Figure 2-35 plots. What if you constrained to sites active in both years and plotted "average % change: 2010-2020" (or is it 2011 – the map label and figure caption don't agree). There doesn't really seem to be much change in the CASTNET dry reduced N (NH₃) between 2000 and 2019, so does that imply that the increase in wet NH₄ from 2000 to 2019 is due to more efficient wet removal of gaseous NH₃

(presumably near sources), does much less conversion to aerosol phase contribute to that, and can you explore and tell that story a bit better?

P. 2-50, lines 12-32: The description of what emissions reductions were modeled for Figures 2-43 and 2-44 seems unnecessarily vague. The figure captions refer to implementation of existing rules & regs by 2032. Does this include attainment of the current annual PM_{2.5} standard of 12 ug/m³ (and what other existing regs)? Does it also include the other expected SO_x and NO_x reductions from EGU retirements mentioned in the PM REA? Could you at least summarize the estimated % national reductions in SO_x and NO_x emissions for the modeled scenario (as well as any reductions in NH_x emissions that might have been included)? Also, while you say there's considerable uncertainty in future reductions in deposition associated with any revisions to the annual PM_{2.5} standard, I assume you included modeled estimates of attaining lower annual averages of 9 or 10 ug/m³ PM_{2.5}, or if not could comment on what additional % reductions in SO₂, NO₂ or NH₃ might be required if the currently proposed PM_{2.5} NAAQS were promulgated. I'm not sure there are great control options for primary (or secondary) OC emissions, but don't really know that. You must have some ideas from the last PM NAAQS review.

Also, just curious why you present these results for Class 1 area sites only? This is OK, and these likely include some of the most sensitive ecosystems and are also responsive to regional reductions in SO_x, NO_x or NH_x emissions – but maybe some explanation of why you present results this way. Presumably these are model results and you could show expected changes everywhere. Also, focus on the “protected” Class I sites while appropriate, misses other sensitive areas (Adirondacks, etc.) and also leaves out large mostly rural sections of the country.

P. 2-53, line 32: The Li et al. PNAS paper needs a date (2016).

P. 4-4, lines 23-25: This effect of continuing but lower than historical deposition needs to be described in more detail. Is it slowing or altering the recovery? What would be the consequence if SO₂ and NO₂ concentrations everywhere were allowed to increase up to the levels of the current secondary standards?

P. 4-5, line 33: Should this read “SO_x, NO_x and NH_x emissions”. Note that SO_x would also include primary sulfate emissions.

P. 4-7, line 23 – P 4-8 line 9: This summary makes it sound like all effects were caused entirely by much higher past deposition, and that full recovery will eventually occur everywhere if today's current lower deposition rates continue. What about referring to effects from “historical and continuing” acidic deposition and discussing how the pace of chemical and biological recovery has been impeded or in some cases precluded by continuing lower but still injurious rates of S and N deposition?

P. 4-27, section 4.2.3.2: Somewhere here you might include some discussion of how deposited SO₂ or SO₄ is converted to sulfide – to what extent, where and what concentrations are toxic.

P. 5-19, lines 17-35: The decision to disregard potential acidifying effects of (oxidized and reduced) N is a departure from the previous S/N NAAQS review. I think you need to present here some clearer illustration of the justification beyond just “see appendix...” While N deposition has declined, S deposition has declined even more (Figs 2-31 & 2-32) and N deposition exceeds S deposition at most sites (figs 2-38 & 2-39). The analysis presented in appendix 5A.2.1 does seem to show that adding N to S would not increase the number of CL exceedances by much in recent years, but it also seems to show that considering N would not have made much difference 20 years ago. Could you tell us something

about those relatively few waterbodies where N dep had or had contributed to acidification? I don't get how N has become less important over time and would like to see some indication of why it was considered in the previous NOx/SOx NAAQS review, but not this one.

P. 5-21, Figure 5-6: This is a relatively poor quality (low resolution) graphic. In areas where sampled waters are close together, its not clear whether the dark gray dots are covering up the light grey dots or vice versa. The dots seem to almost completely hide the “likely naturally acidic” areas along the mid-Atlantic coast. Here and for several analyses that follow, it's not clear what your total population of sampled lakes represents. Does the NCLD database include results for 14,000+ waterbodies concentrated in areas that are acid sensitive, primarily, the eastern U.S. and the Rocky Mountain and Pacific Northwest regions of the west?” If so, what are the grey dots in Fig. 5-6 that lay outside these regions?

P. 5-22, lines 12-23: Again, it's not clear what this population of 13,824 lakes with CLs represents (lakes expected to be acid-sensitive, lakes with ANC < 200, what?). Are the percentages in Table 5-1 relative to the 13,824 lakes or to all US lakes? What about also presenting the numbers of lakes (does 4% = about 550 lakes?). How does 13,824 relate to the total # of US water bodies?

Figures 5-7 through 5-10 are very impressive! With such a dramatic reduction in the # of waterbodies exceeding their CLs over the 20-year time period of your analysis, is there something interesting you can show us about what happened in that subset of lakes that once exceeded and more recently do not exceed their CLs? How many waterbodies exceeded in 2001-3 and how many fewer in 2018-20 (and what do you project by 2032? Can you summarize the chemical changes that took place over that 20 yr period in those “chemically recovered” lakes— compared to the associated changes in TDEP?

PP. 5-33 to 5-34: With all this discussion focused on S deposition “thresholds” at which various changes occur, would it be helpful to show TDEP maps – for the most recent and perhaps also for the 4 time periods – which are not the smooth gradient maps, but rather which have distinctly colored areas better showing the lower ranges of S dep referred to in the text on p 5-34?

P. 5-37, Figures 5-14 & 5-15: Since the “qualifying” ecoregions are sort of self-selected as regions with high #s of CLs, which are in turn regions where acidification effects have been of concern, I have a hard time processing what a “% of waterbodies” (or a “% of ecoregion-time periods”) actually means. Isn't it an indicator of variability within ecoregions, or an indicator that maybe the ecoregions weren't the best idea? Would it at least be possible to provide a few text links between % of waterbodies and # of waterbodies. For example, in describing Figs 5-14 and 5-16, you could include a statement like: “For the most recent 2018-2022 time period, there were x waterbodies exceeding their CL for an ANC of 50 ueq/l and y waterbodies exceeding a CL at 20 ueq/l.”

PP. 5-39 to 5-41: I assume these are the 25 ecoregions on which previous analyses were based. Would the maps look more interesting if you added one more category – 0-5% and 5-10%? Also, would it be possible to include parentheses in the legend indicating how many waterbodies are included in the 0-10% category, or maybe just use dots to show the lakes that exceed.

P. 5-44, lines 4-13: This case study analysis would be more informative if you also reported the estimated S TDEP for each region, perhaps for the 5 time periods. Another potentially useful kind of information from the historical data might be to look for the time periods at which previously exceeded CLs were attained and/or to look for any signs of chemical or biological recovery in individual

waterbodies which once exceeded and have now attained their CL in these case study areas. Surely, there must be waterbodies within these case study areas (or elsewhere like the Adirondacks) where you have historical lake chemistry data (SO₄, ANC, AL, BC) that you could contrast with declining SDEP over your 5 time periods. Show us how the lake chemistry responds to dep changes.

Would it be possible to consider similar case studies for N deposition, perhaps using the N CLs the National Park Service has identified for its sensitive Class I areas?

P. 5-47, line 3: Could you add a parenthetical indicating the number of waterbodies that 4% would represent. Would it also be reasonable to project how this number might be expected to change in the future if the future S reductions through 2032 that you mention in chapter 2 are achieved?

P. 5-49, Section 5.3.2: Are N deposition contributions to eutrophication of marine estuaries no longer an issue? TMDLs for Chesapeake Bay, Long Island Sound/ CT River...?

P. 5-50, lines 2-4: Need a bit more explanation of what “false negatives” means here.

P. 5-66, lines 10-29: The significance of this detailed description of “percentage of ecoregion-time periods” is difficult for me to grasp. I would be more interested in hearing about what happened chemically (and biologically) in the waterbodies that formerly exceeded their estimated CLs and have now attained them.

P. 5-66, lines 28-29 and 32-36: Aren’t current S Tdep rates below 5 kg/ha everywhere? It’s really hard to tell from your only map in chapter 2. You could at least explain in the text how these dep rates described here – 13, 9-10, 6-9 kgS/ha/yr - compare to current dep rates in the study areas.

P. 5-69, lines 29-34: Is it clear that these changes resulted from use of a newer, better model, rather than from newer, lower rates of deposition in the more recent studies?

P. 5-70, line 20: This is just one example, of which there are many, where there’s a reference to information we’ll see (clearly) much later in an appendix, without a clear indication of what the future information will show. If you have some good, illustrative tables or graphs, why not include them in the earlier summary chapters.

P. 5-70, lines 30,31: Maybe a footnote explanation of what “functional type grouping” means.

P. 6-3, lines 11-13: Was it or would it be necessary to spatially average the transference ratios across an entire ~ 10,000 km ~ ecoregion? Couldn’t you calculate and use the TR at each 4km grid cell for which you have all the TDEP components and CMAQ model results. Use the (few) sites where you have sufficient measurements to evaluate the reasonableness of the gridded TR calculations. By the way, whatever became of the proposed “pilot network” EPA planned to set up to test the proposed measurement metrics? Another possible approach: there’s nothing sacred about the Ecoregion level III areas. You could break them into smaller pieces if model results indicate high spatial variability in TR estimates.

P. 6-4, line 6: You could also mention protection of “air quality-related values” as it includes other deposition-related effects more directly related to the current NAAQS review. See for example Fox et al. (1989), Adams et al. (1991), US Forest Service et al. (2010).

P. 6-6, Fig. 6-2: The “Class 1 areas” is a nice idea, but clearly limits the coverage in the East where concentrations, deposition and acidification effects have been greatest. There are several other eastern “IMPROVE Protocol” sites that I think meet your criteria. Bondville, IL; PMRF, VT and maybe others. Also, I’m surprised you go all the way back to 1988, which includes much higher concentrations and depositions, with relationships that may have changed over time – especially for N. Also, 1988 assures varying start dates among your sites. For example, ACAD, GRSM and Everglades didn’t start ‘til 1999, Voyagers in 97.

P 6-7, lines 18-33 and Fig. 6-4: These scatter plots are interesting, and I realize you can only look at so many comparisons, but a few that seem relevant but not examined might include

- IMPROVE N (which should include both measured N from NO_3 and assumed N from NH_4 that would be required to neutralize SO_4 and NO_3) vs. NADP N.
- IMPROVE NH_4 (estimate as indicated above) vs. NADP NH_4 .
- Might there be a non-mass $\text{PM}_{2.5}$ indicator like $\text{PM}_{2.5}$ sulfate + nitrate + ammonium or PM N, S or N+S – or maybe $\text{PM}_{2.5}$ mass minus carbonaceous matter minus crustal minus seasalt?
- In the previous NAAQS review, weren’t you considering “total atmospheric S” (sum of PM SO_4 and Gaseous SO_2), and “total atmospheric oxidized N (PM NO_3 + NO_y)”?

P. 6-11, lines 6-17: Possibly also S and NO_x aerosol conversion rates have strong and opposite seasonal patterns. Both also have summer-peaking trends in deposition velocities, so NO_x dry deposition is >> than SO_x in summer.

P. 6-12, lines 8, 9: Can you give the 21-yr time period for the Zhang et al. modeling? I’m wondering how well it compares to the 1988-2018 period of the measured data, and if constrained to reflect varying measurement time periods among sites.

P. 6-23, lines 12-15: Another example (there are many) of how the reader really has no idea what you’re describing without jumping to the appendix. In some cases, maybe this could be fixed by adding a bit more description and a graphic example or 2 into the chapter. Another idea might be to include a “reversible hyperlink” (no idea if such a thing exists) which would take the reader to jump to the relevant appendix section and add a “return to original location” button.

In this case, I still don’t fully understand the trajectory-based EAQM calculations described in the appendix. Off hand, 48-hr trajectories seem a bit short given lifetimes of sulfate & nitrate aerosols are more like 3-5 days. Your illustrated example, Figure 6A-1 surprises me that such a few monitors such a short distance outside the ecoregion are relevant. I wonder if this is related to selection of a $\text{PM}_{2.5}$ indicator and/or an ecoregion with relatively high local emissions? Would a similar image result for SO_2 or NO_2 monitors outside a more remote ecoregion?

P. 6-24, Figure 6-25: Wow!! This really surprises me, especially given the predominance of western sites in your analysis and likelihood of strong influence of organics from both summer forest fires and winter woodstoves on $\text{PM}_{2.5}$ mass at those sites. Maybe the relatively high elevation IMPROVE sites are mostly immune from winter (valley) woodsmoke and maybe the summer forest fires don’t have huge influences on annual average $\text{PM}_{2.5}$. Or wait, maybe forest fires are actually important sources of NH_4NO_3 . See for example Paulot et al. (2017)²¹, Kopplitz et al., (2021)³⁴ Campbell et al. (2022)²². I don’t really think that’s it. Whatever it is, I really like what you’ve discovered and illustrated here, even if I don’t quite believe it. While the colors are hard to distinguish, it doesn’t seem like high correlation is

driven by past conditions when S and N represented higher fractions of PM_{2.5}. If anything, the fit seems even tighter looking at the more recent, cleaner years. This is strange!

P. 6-36, lines 7-20: In drawing these conclusions, you only consider SO₂ as a potential indicator that doesn't (currently) seem to relate very strongly to S deposition. Other potential legit indicators that would correlate better might include: aerosol SO₄, S or total atmospheric S (from SO₂ and SO₄). Is there any reason these metrics couldn't be used as an indicator?

P. 6-40, lines 7-22: Same comment as above. Why are you constrained to SO₂ – only?

P. 6-41, line 8: You could rephrase this to "... reduced nitrogen (e.g. NH_x) ..." Not entirely clear what the intended meaning is here, but an alternative statement might be "Additionally, the results suggest that in most areas reduced nitrogen (e.g., NH₃, NH₄⁺) contributes more than half of total N deposition, with higher contributions in areas near emission sources of NH₃."

P. 6-41, lines 17-28: Non-S, non-N components of PM_{2.5} mass (especially organic matter from forest fires and crustal material from dust storms) generally tend to be more episodic than S and or N components, so the geometric mean might be a better, more stable PM_{2.5} indicator than the arithmetic mean. Also, for application of the Regional Haze Rule, clever methods have been developed to screen out episodic contributions from forest fires and dust storms when considering the most (anthropogenically) impaired days. Similar screening might also be applied when considering a PM_{2.5} mass or PM_{2.5} species indicators. Recent analyses suggest that wildfires can be important contributors to deposition of N and to a lesser extent S, especially in the Northwest (Kopitz et al., 2021).³⁴ However, it may arguably prudent to intentionally exclude influences of such (non-controllable) sources from a regulatory metric.

P. 7-7, lines 18-20: I think this "relatively greater role of S under more recent air quality conditions" needs to be demonstrated more clearly in section 5.2 (I can't seem to find the indicated section 5.2.1.4). Also, the analyses presented in Appendix 5A.2.1, while showing relatively slight increases in CL exceedances when both S and N deposition are considered, compared to considering S only, doesn't really seem to show that this difference has been increasing in recent years. That is, by these calculations it looks like N contributions were about as unimportant in 2000 as they were in 2020.

P. 7-10, lines 1, 2: Could you add something like "These 550 (or however many) waterbodies are primarily distributed in (which) ecoregions and represent an improvement from the (however many) waterbodies for which critical loads were exceeded in the earlier 2001-03 time period." The "4%" figure reminds me of David Stockman (Regan's Budget Director) wondering in 1980 "how much are the fish worth in the 170 lakes that account for four percent of the lake area of New York? And does it make sense to spend billions of dollars controlling emissions from sources in Ohio and elsewhere if you're talking about very marginal volume of dollar value, either in recreational terms or commercial terms?" (Boyle, 1981)²³

P. 7-10, lines 8-25: Again, hard for me to get the significance of this analysis. What if you had to summarize at the end of this paragraph in a single sentence, starting with "In other words..."? Since, you have a fairly large population of surface waters which used to exceed their (estimated) CLs, at various dates and at various threshold levels, what about summarizing the changes in chemistry that have taken place in those theoretically "recovered" waters (maybe compared to the group of 550 that still exceed)? Was the target ANC achieved, did Al decrease, did DOC increase, any biological changes?

P. 7-13, lines 7-9: This doesn't seem to make sense. First you say that SO₂ tends to deposit closer to sources and SO₄ farther away (true enough). And then you say that in the West where S is lower, S deposition may come more equally from SO₂ and SO₄. This is sort of a double non sequitur. You didn't say anything about less equal distribution of deposition forms in the East, nor how lower emissions would tend to affect the partitioning.

P. 7-15, line 26 – P. 7-16, line 11: You don't really give much actual consideration to these potential alternative metrics. There are about 150 sites each in the IMPROVE and CSN networks (about 300 total counting supplemental), that employ standard methods to collect samples and analyze for SO₄ and S (redundant), with several collocated intercomparison sites. These represent nearly as many existing sites for PM SO₄ (and NO₃) as there are in the existing SO₂ and NO₂ networks, and measure more regionally representative pollutants that relate more directly to deposition. PM_{2.5} S, N or the sum of PM_{2.5} S+N would likely be a better indicator for both S and N deposition than would SO₂, NO₂ or PM_{2.5} mass. As an indicator for S deposition, yes SO₂ + SO₄ would likely work best (as proposed in the last NAAQS review – when you also proposed setting up a pilot network to test its efficacy [along with some proposed total atmospheric NO_y measures]). But as you observe, there aren't many CASTNET sites, the number is shrinking, and the pilot network...

P. 7/21, line 26: You could change "...inorganic nitrogen (e.g., NH₄) contributes..." to "...reduced nitrogen (e.g. NH_x) contributes..."

P. 7-22, lines 4-10: A tighter secondary PM_{2.5} NAAQS would have the additional benefit of protecting visibility. If there is concern about the non-S&N components of PM_{2.5}, the sum of S + N from roughly 300 PM_{2.5} speciation sites would be a better indicator of both N and S deposition and would also be an excellent indicator for (anthropogenically impaired) visibility effects. This metric would most directly focus on the species of concern, would include influence from NH₄NO₃ lost from Teflon PM_{2.5} filters, and would emphasize the potentially controllable components of PM_{2.5}.

P. 7-25, lines 16-23: This observation that injurious levels of S deposition were observed in past years when SO₂ standards were attained, also illustrates that SO₂ emissions were substantially decreased for reasons other than attaining the SO₂ NAAQS. I suspect that most of the substantial reductions in both SO_x and NO_x emissions in the past 25 years have been driven largely by efforts to attain the PM_{2.5} NAAQS (or from control strategies where estimated benefits were largely justified by reductions in PM_{2.5} at levels above and below PM_{2.5} NAAQS). This (successful past track record) seems like a major point in favor of considering a PM_{2.5} mass or PM species indicator to reduce future N and S deposition.

Since organic matter is becoming the predominant mass fraction (and will be more-so in the future) what about considering a proposed metric called something like "non-carbonaceous PM_{2.5}"? This could theoretically be "measured" relatively inexpensively by applying (non-destructive) FT-IR analyses to PM_{2.5} FRM (or other PM_{2.5}) filters. The FT-IR analysis could also provide hugely valuable information about both the mass and (source related) chemical composition of PM_{2.5} organic matter which is not currently well-characterized by current STN and IMPROVE measurements – which just measure so-called "OC" and apply some fudge factors. This could be a great indicator which would be more relevant to S and N deposition than PM_{2.5} mass – and would provide much-needed info on carbonaceous PM. FTIR also has the potential for quantifying SO₄, NO₃ and maybe NH₄ from Teflon filters currently used to measure PM_{2.5} mass Probably not ready for prime time but could be a useful research need.²⁴⁻²⁸

P. 7-27, lines 10-14: This seems like a good idea. Reminds me that it would be useful here or in chapter 2 to see a bit more analysis of how the pollutants vary over the various averaging times – like maybe some scatter plots comparing 1-hr and 3-hr max vs. annual average SO₂; 1-hr vs. annual NO₂, etc. It looks like maybe only 2 sites would exceed an annual SO₂ average of about 8 ppb over the past 3 years – and they likely exceed the primary, such that a much tighter secondary wouldn't really cause any pain, and could be very useful to prevent backsliding. Conceivably, you could go even lower, with the annual mean (like the 1971 annual PM secondary) “as a guide to be used in assessing implementation plans to attain” the primary, and to prevent future ecological damage.

P. 7-30, lines 15, 16: It's not exactly clear where this 15 to 12 range comes from – other than that it spans the difference between the current secondary and primary standard. As previously mentioned, the current secondary standard set at a higher level than the primary protects nothing, it has no effect and could be discarded. It wouldn't really add protection (of visibility or eco effects) to lower it to 12, but it would look a lot less silly. Since a lower primary annual PM_{2.5} NAAQS to a 9 or 10 has recently been proposed, wouldn't it make sense to consider lowering the secondary to a similar level? It would cause no additional nonattainment and could be justified for protection against both eco effects and visibility.

P. 7-30, lines 24-37: Since no site has averaged greater than 40 ppb for the past 20 years, or above 35 for the past 15 years, it seems safe to assume that lowering the annual NO₂ to somewhere between 53 and 40 ppb wouldn't actually do much (but could potentially prevent backsliding). Also seems unlikely that this would cause “appreciably greater uncertainty”.

P. 7-30, line 32 to 7-31, line 6: I think you maybe overstate the profound uncertainties here. We've got > 20 years of speciated PM data from IMPROVE, CSN & CASTNET networks + NADP and TDEP data. We understand very well what the space/time patterns and source/receptor relationships are for S and N emissions concentrations and deposition, and we have the clear evidence that changing S and N deposition relates to changing ambient air concentrations and results from changing emissions of S and N. PM_{2.5} S + N, would be an excellent indicator, which would relate more directly to S and N deposition – especially in the future, and would also focus on PM components which are most unambiguously anthropogenic (controllable).

You have repeatedly emphasized the uncertainties (400 times) about almost everything. Typically, uncertainties are used to justify inaction on a NAAQS (lest we unnecessarily burden the polluters). Uncertainty could also work the other way around. Given the (what millions?) of plant, animal and fungal species on the planet, the incomprehensible multitudes of ways in which they interact in different ecosystems, the limited research, etc. and the huge remaining uncertainties about the consequences of altering our/ their chemical climate – doesn't it make sense to err on the side of caution (lest we unnecessarily burden the environment)? Since current secondary SO₂ and NO₂ NAAQS are attained everywhere, lowering or otherwise revising a secondary NAAQS for these pollutants, or for PM_{2.5} (preferably PM species) as a more comprehensive indicator to concentrations near to or reasonably below current levels would be consistent with accommodating the uncertainties in both directions.

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Chapter 6 – Relationships of Deposition to Air Quality Metrics

To what extent does the Panel find the investigation of relationships between air concentration and deposition to be technically sound and clearly communicated?

- Chapter 6 aims to establish relationships between ambient concentrations of S and N oxides and PM_{2.5} and the deposition of S and N to ecosystems, with the goals of showing how changes in emissions/concentrations affect deposition and determining whether the current secondary standards are protective of public welfare.
- The approach, which involves correlating a variety of metrics from various monitoring networks, seems reasonable and the intent is clearly communicated.
- **Terms/abbreviations:** It would be helpful if certain terms/abbreviations were used consistently throughout the text. For example, the text states that the IMPROVE network measures the sulfate and nitrate components of PM_{2.5}. These components are referred to variously as SO₄²⁻, NO₃⁻, nitrate PM_{2.5}, and sulfate PM_{2.5}.
- **Scatterplots:** Page 6-15: The text notes that ammonium is included in total nitrate in the CMAQ model (in contrast with IMPROVE and CASTNET). Differences between CMAQ, IMPROVE, and CASTNET in the species that comprise the variables used in the scatterplots need to be clarified earlier in the chapter to aid in interpretation. For the Class I areas analysis, a table similar to Table 6-2 with the definitions of “total nitrate”, “total sulfur”, “wet dep N”, etc. would be helpful.
- Figure 6-10: How is “total deposition” defined here (S + N)? What is IMPROVE N? These terms were not defined earlier in the text.
- The change in labelling among the various scatterplots makes them hard to interpret. How is IMPROVE N defined (NO₃ PM_{2.5} + NH₄ PM_{2.5})? On page 6-7, it states that NH₄⁺ is not measured by IMPROVE, therefore this is confusing. Page 6-20 answers the above questions. Text on lines 4-7 should be moved to earlier in the text for clarity.
- Page 6-8: The weak relationships between wet N deposition and nitrate and between wet N deposition and the sum of nitrate and nitric acid support the role of ammonium in driving N deposition. This point could be made more strongly at the end of this section.
- The scatterplots show that wet S deposition is weakly correlated with IMPROVE nitrate PM_{2.5}, however wet N deposition is strongly correlated with CASTNET total sulfur. What is the explanation for this?
- Why is it assumed that most of the deposition to Class I areas, which are largely situated in the western US, is from wet deposition? On Page 6-4, the text notes that dry deposition contributes 57% to total deposition for N and 40% to total deposition for S at these sites. Depending on aridity and elevation and other factors, the proportion of dry deposition is likely to vary widely.

- **Impacts on urban ecosystems:** Chapter 6 focuses on concentration-deposition relationships for remote areas. While these areas are of ecological and legal importance, it would also be useful to conduct a separate analysis for urban areas. In recent years, there has been a proliferation of studies focusing on atmospheric deposition of N and PM (and its components) in urban areas (e.g., Barrett et al. 2019; Decina et al. 2019; Conrad-Rooney et al. 2023). For example, using data from the NADP NTN network, Conrad-Rooney et al. (2023) found elevated rates of ammonium, inorganic N, and sulfate wet deposition to urban compared to rural sites, while a global analysis by Decina et al. (2019) showed that N deposition was approximately two-fold greater in urban compared to rural sites.
- The adverse effects of atmospheric deposition on urban ecosystems are of critical importance to public welfare given that most of the U.S. population lives in urban areas and derives benefits from the ecosystem services provided by urban nature. These adverse effects are also important to consider in the context of existing disparities in air pollution in U.S. cities (Chambliss et al. 2021). Within cities, it is well established that areas near roads and downwind of industrial sources experience elevated levels of air pollution and PM deposition (Karner et al. 2010; Ponette-González et al. 2022). Because communities of color often live in said areas, these communities are likely to not only experience the disproportionate effects of air pollution but also the associated effects on urban vegetation and ecosystems. The availability of concentration and deposition data from monitors in urban and suburban areas could facilitate such analyses.

What are the CASAC views on the consideration of key limitations and associated uncertainties?

- One key limitation of the analysis for Class I areas is presented. The data are largely for sites in the western U.S., which contrasts strongly with the eastern and central U.S. in terms of emissions, air quality, meteorology, and terrain.
- Another limitation for the Class I areas analysis is the reliance on wet deposition measurements as a proxy for total deposition. In arid and montane regions, total deposition rates are highly variable across space and time due to complex topography, meteorology, and emissions (Weathers et al. 2000, 2006). Moreover, estimates are uncertain due to the challenges associated with measuring atmospheric deposition in these locales (Fenn et al. 2009).
- The second limitation identified is the HYSPLIT modeling approach and the uncertainties associated with the modeling. Rather than generating a new approach, building on a previous approach would likely lend confidence to the modeling. Although some details are included in the Appendix, it is not clear how the various parameters were determined (e.g., plume height, percentage of hits, etc.) and/or if sensitivity analyses were conducted.

Corrections to Chapter 6

- I have several concerns about the results presented in the memo. As presented, my primary concern with the relationships derived from the EAQM approach, is that the correlations between NO₂ and PM_{2.5} and nitrogen deposition are somewhat weak for eastern ecoregions, very weak to nonexistent for western ecoregions, and not representative of the entire country. This leads me to question the utility of the results as they would seem to suggest that standard setting would be based solely on relationships for eastern ecoregions.

- The memo does not indicate whether the correlations presented are statistically significant.
- Correlation coefficients are not presented for the relationships between SO₂ and PM_{2.5} and sulfur deposition. There are also no figures of this revised analysis.

Editorial suggestions

- Page 6-1, Line 16: “are set in terms of an ambient atmospheric concentrations”. Cut “an”
- Page 6-2, “Thus, as the chemical and physical forms of nitrogen and sulfur vary in the atmosphere, it leads to differences in the rate of deposition, and causes variability in the relationship between total air concentrations and atmospheric deposition.” Rewrite, awkward sentence.
- Page 6-4, Line 6: Delete “as well” at end of sentence.
- Page 6-10, Figure 6-5: The acronyms in the scatterplots are not defined in the legend and do not appear in the text before the scatterplots are presented. For example, Page 6-7, Line 25: where the text says CASTNET total nitrate (HNO₃+NO₃⁻), additional text could be added to clarify (HNO₃+NO₃⁻, CASTNET TNO₃ in Figure 6-5). It would be helpful to define CASTNET TS in the text (SO₂ + SO₄²⁻).
- Page 6-22, Line 29: Delete ‘s’ from areas.
- Page 6-42, Line 22: Phrase needs to be rewritten “Although, ...”

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Chapter 7 – Review of the Standards

What are the Panel views on the discussion of key considerations in sections 7.1 and 7.2 and preliminary conclusions?

- Chapter 7 presents options for consideration to better control for N and to some extent S deposition. Additional text on how these levels were determined is needed.
- Based on the analyses presented, the preliminary conclusion that the existing SO₂ standard provides protection against the direct effects of effects of SO_x in ambient air is sound.
- In contrast to the standard to protect against the direct effects of SO₂ in ambient air, a new standard to control for the adverse effects of S deposition is supported by the body of evidence reviewed in the policy assessment.
- Chapters 6 and 7 underscore the finding that the relationship between SO₂ and S deposition is stronger when a standard with a longer averaging time is used. The uncertainties associated with revising the standard are also clearly presented.
- One option presented to address deposition-related effects of S is to revise the existing standard to a level of 10-22 ppb SO₂ averaged over three years. This evidence is based on the EAQM approach, which shows that “*median S deposition in the Ecoregion III areas is <5 kg/ha-yr when the annual average SO₂ concentration, averaged over three years, at contributing monitors was less than 22 ppb and the majority of monitors were below 10 ppb*” (Page 6-36). Figure 6-18, to which this text refers, indicates that the data are “from the contributing monitor with the maximum value for each ecoregion”. On page 6-36, this should be clarified (i.e., that this is from the monitor with the maximum value for each ecoregion), and the figure should be cited (Figure 6-18). It would also be worth highlighting as well that the annual average SO₂ design values (ppb) at U.S. sites have been below 15 ppb at the vast majority of sites since 2018 (Figure 2-25).
- To address N-deposition related effects, one option presented is to revise the level of the annual NO₂ standard to <53 ppb to as low as 40 ppb. Given the data presented in Figure 2-21, which show that annual mean NO₂ concentrations (ppb) are much lower, on average, than this, especially in the last

10 years, this needs to be lower or better justified. What are the 3-year average N deposition values associated with such levels?

- The results in Chapter 6 suggest that the PM_{2.5} annual average is strongly correlated with and could be used to control for N deposition. I agree with EPA that contributions of other components to total mass and changes in composition across space introduce uncertainties here. Additionally, changing disturbance regimes across the U.S. (e.g., dust, fire) and associated shifts in PM composition both now and in the future might affect the usefulness of such a revised standard.

What are the CASAC views regarding the areas for additional research identified in Chapters 3 and 4? Are there additional areas that should be highlighted?

- The document identifies several areas for additional research that would facilitate future assessments.
- The current policy assessment is based on the 2020 Integrated Science Assessment, which includes a review of the literature through early 2017. Seven years have elapsed since that time, and thus the current policy assessment does not capture a growing body of research on PM deposition to ecosystems and corresponding ecological effects.

Ecological effects of PM deposition

- In addition to the Grantz et al. (2003) paper (cited in the ISA), there is a more recent review of PM deposition effects on plants in Burkhardt and Grantz (2017). Aside from this review, there are a few studies not covered in the ISA, many conducted since 2017, that demonstrate the effects of PM deposition on vegetation, which include wax degradation and decreased drought tolerance (Burkhardt et al. 2018) and decreased plant growth (Locosselli et al. 2019) (tree decline). Research on the effects of elemental carbon deposition to leaves indicates that EC can penetrate leaf stomata (Tao et al. 2022) and affect the concentration of photosynthetic pigments (Byčenkienė et al. 2022). Deposition of PAHs to leaves has been shown to alter ecohydrological processes as well. Klameraus-Iwan et al. (2018) showed that PAHs decreased canopy water storage capacity of trees. Further research is needed on the deposition-related effects of PM and its constituents, such as black carbon. More information is needed on how deposition affects leaf temperature, photosynthesis, transpiration, and soil processes. These effects are important given their public welfare implications (i.e., potential for decreased productivity and carbon sequestration, increased stormwater runoff, and mortality).
- Changing wildfire regimes and changes in PM deposition: Further attention to the ecological effects of PM deposition is especially relevant in wildfire-prone areas and under changing fire regimes.

Editorial suggestions

- Page 7-4, Line 21-22, “With regard to direct effects of PM in ambient air, the information on ambient air concentrations is well in excess of the existing secondary standards”. This sentence is not clear to me.

- Page 7-13, Line 31-33 – The second half of this sentence could appear at the beginning of Chapter 6 to clarify the goal of the analysis.

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Dr. Stephen E. Schwartz

During the September 5-6, 2023, public CASAC meeting, I found Dr. Boylan's presentation of alternative standards rather compelling. Therefore, I joined in with his dissent. However, after the meeting, and upon further consideration, I am withdrawing from his dissent. My preference would be to not recommend any alternative standards until the agency develops a 2nd Draft PA for CASAC review.

As outlined below *I do not consider EAQM (or any other long range transport model) necessary or even relevant to the standard setting process.*

It is my understanding that the intent of the document under review is to propose secondary AQ standards for S, N, PM, and to justify them.

That said, I think such standard setting could be done much more succinctly and with much more solid justification than in the draft report. For example a document such as the one under review is not the place for presenting scatterplot matrices of this variable versus that, figures 6-4 and 6-5. The place for that is the peer reviewed literature. As elaborated below, a document such as the one under review would refer to the findings of the pertinent peer reviewed articles showing that there is this or that good correlation that serve as the basis for standard setting, rather than to show such scatter plots de novo in the document and have CASAC serve as the peer reviewers.

I would say the same thing about the EAQM. There are several peer reviewed models in more or less widespread use. If it is necessary to introduce another one, then this document is not the place to do that. The place to do that is the peer reviewed literature, with suitable comparisons of model and observations and everything else that goes with giving credence to a model of this sort (or any sort).

However, beyond the issue of *where* to present the model, I would say that *modeling transport and deposition on the hundred km scale or multi hundred km scale is simply not relevant to standard setting.*

Part of the issue that seems to have motivated the modeling is that the standards must be (at least as is my understanding) couched in terms of concentrations, even though the objective is to limit deposition to protect sensitive ecosystems. Hence the argument is made that the species under consideration, sulfate, nitrate, are derived from emissions hundreds of km away. But to my thinking that is irrelevant. It is conceded in TDM that dry dep of gases is local and can be calculated with sufficient accuracy using deposition velocity approach. I think it is also conceded that dry dep of aerosol sulfate and nitrate is small relative to wet dep and hence of very small concern.

The issue then is wet dep of sulfate and nitrate, for which the argument is made (correctly) that these species are produced during multi-hundred km transport from source to receptor. But that is, to my thinking, *no reason or justification to model this transport.* Rather, I would argue, the sulfate and nitrate that is deposited in precip results from sulfate and nitrate (and nitric acid) that is present at the locus of deposition (or within, perhaps a few km, the distance that the storm travels during the time that it takes for the precip to reach the ground from the cloud generating the precip). So, again to my thinking, the sulfate and nitrate are local, albeit produced from emissions at distant sources. Hence this is justification to set a standard that is based on local sulfate and nitrate, and certainly offers no reason to do long range transport modeling.

This view is strongly supported by the panel in Fig 6-4 showing strong correl between sulfate wet dep and conc. The inverse of the slope of the line in the panel row 6, column 5 would give an observationally determined wet deposition velocity, $(\text{kg ha}^{-1} \text{ yr}^{-1})/\mu\text{g m}^{-3}$, amount area-1 time-1 / (amount volume-1) = length time-1. To my thinking that provides the desired relation between conc and dep for sulfate. I underscore that that relation is *based on local measurements, with no requirement for a long-range transport model*. [I think that the reason that the plot for nitrate fails is that it does not account for nitric acid.]. The tight correl for sulfate lends strong support to such an observation-based approach. The document, page 6-11, notes the strong correl betw sulfate conc and sulfate dep, suggesting that that relation can be used in standard setting without use of long range transport models. The same para suggests why the correl is weaker for nitrate, likewise suggesting the role of nitric acid.

Subsequent to the public meeting I examined the result shown in the scatter plot matrix Figure 6-4 of concentration of sulfate in air versus wet deposition of sulfate. (In the first draft provided to the panel the values of the quantities plotted were not legible but this was corrected in the document NOxSOxPM-PA-Draft-ERD_finalv3.pdf) Based on my examination of that figure I was able (making I hope correct assumptions of the units of air concentration and wet deposition amount) to calculate a wet deposition coefficient for sulfate based on the slope of that panel, which I took to be $32 \text{ kg SO}_4 \text{ ha}^{-1} \text{ yr}^{-1} / 6 \mu\text{g SO}_4 \text{ m}^{-3}$. I took the deposition velocity to be the slope flux/concentration, analogous to the familiar dry deposition velocity. This analysis came up with the value of wet deposition velocity $1.85 \times 10^{-3} \text{ m s}^{-1}$, which translates to 1.12 km per week. That is a very reasonable number give typical sulfate lifetime of about a week and typical boundary layer height of 1 km.

I emphasize that to my thinking it is not the job of the CASAC to do this kind of analysis for EPA. Rather that sort of analysis should have been done in the peer reviewed literature and the results presented as part of the basis for standard setting to relate dep flux to conc. I am afraid CASAC got hung up in the details of these plots instead of simply telling EPA to go back and publish these findings in the peer reviewed literature, draw the conclusions that are appropriate for standard setting and use them in the standard setting process.

To my thinking such an approach on the part of EPA would result in a document smaller than the present one by at least a factor of two, and present a much stronger basis for standard setting.

My overriding concern over the document as presented is that it is too little of large picture and too much of detail. I would like to see big-picture questions and answers. I raised some of these questions in the meeting with EPA staff, some of which were answered, and some are being raised de novo here. I hope EPA staff would consider these questions and respond to them.

1. Do the standards have to be uniform nationally? I understand the answer to be yes.
2. Are the standards restricted to air concentrations (as opposed to, say, deposition fluxes). Again, I understand the answer to be yes.
3. Are the present concentration standards (for NOx, SOx, PM) being met? If not by how much are they being exceeded (absolute and/or fraction). If they are being exceeded is there a strategy to achieve these standards? How has this strategy been developed and tested? What is the uncertainty (for example as a probability distribution function) that such a potential strategy would achieve the goal of meeting the standards? The pertinent development, testing, and determination of uncertainty should be presented in the text.

4. Setting aside issues of deposition to sensitive ecosystems, are the present concentration standards sufficiently protective of the welfare issues that the secondary standards are meant to protect? Is there any evidence that these standards are too high or too low, that would lead to recommendations to raise or lower the standards? This evidence should be presented and discussed.
5. Based on deposition measurements, do present deposition fluxes result in harm to sensitive ecosystems? Again, is there any evidence that present deposition fluxes are too high or too low, that would lead to recommendations to set target deposition fluxes that would be lower or higher than present deposition fluxes? Again, this evidence should be presented and discussed.
6. If present deposition fluxes are greater than would avoid harm to sensitive ecosystems is there a strategy to reduce deposition to a rate that would be protective of such sensitive ecosystems? How has this strategy been developed and tested? What is the uncertainty (for example as a probability distribution function) that such a potential strategy would achieve the goal of meeting the such a target deposition rate? The pertinent development, testing, and determination of this strategy and determination of associated uncertainty should be presented in the text.

This is an enormously comprehensive report. I have examined closely Sections 2 and 6, with many specific comments. I have many concerns both with the underlying science and with the presentation and have tried to voice them along the way. I hope that these will be considered in revision of the draft document. In many instances I have asked for clearer statement of definitions or approach. Many of the graphics can be greatly improved.

Figures 5B9 and 10 are enormously important as a testament to decrease in deposition over the 25 year period. EPA should take great satisfaction in this.

To highlight some of my major concerns:

I am surprised visibility is not included in this review. It would seem to be a major component of secondary effects of PM. It is not clear to me why visibility is not included.

PM exerts cooling effect on climate through direct light scattering and modification of reflectivity and persistence of clouds. It is hard (for me) to make a connection between that and public welfare, but perhaps that should be explicitly noted.. I do not think reduction of solar irradiance is sufficient to have an effect on agriculture at least at current levels of aerosol optical depth, but I think that should be explicitly noted. There are exceptions: the recent smoke events (east coast) and prior year (California) had effects on health (not part of sec stds) but also visibility limiting aviation take-off and landing. This would seem to be within the remit of this report, but perhaps not within the remit of EPA, as not something that can be controlled by regulation, but it seems all of this requires explicit discussion.

Figs 2-38, 2-39. It would seem essential to show maps of wet dep and dry dep separately. The sum does not give a sense of the relative magnitudes. Wet dep is interpolated from measmts, whereas dry dep involves interpolated concs times dep velocities, which has inherently more uncertainty. If dry dep is small fraction, then the large uncertainty in dry dep is of relatively little importance, but if dry dep is the large fraction, then that uncertainty looms large.

The "transference ratio" is extensively employed with considerable reference to the ISA report. I think that the present report should define the transference ratio more explicitly and note that it has dimension length/time and hence is a velocity; it should thus be presented as such, in units of cm hr⁻¹ or the like. Also the term "ratio" should be eschewed here; a ratio is a dimensionless quantity, the quotient of two quantities in the same units. "Transference quotient" or even better "Transference velocity" would be much preferred. From a science perspective a transference velocity would at best apply to a single chemical species and only to dry or wet deposition separately. It makes little sense to speak of a transference ratio of SO₂ + sulfate. SO₂ is deposited largely by dry dep; sulfate largely by wet dep. So a ratio of the sum of the concs the sum of the deps has no scientific underpinning and, it would seem to me, should be avoided. See further comments in pdf.

A figure such as Fig 6-7 is potentially quite valuable. There are two impt steps in the cmaq modeling: modeling the conc as a fn of location and time, and then modeling the dep as a fn of loc and time. The quotient is the transfer velocity. So all should be shown. To be sure we don't have the measmt of dry dep flux, so we have to rely on the model, but we need to have some measure of the confidence in the model. The ratio should have units (e.g. cm day⁻¹ or m yr⁻¹) and presented for wet and dry, and it should certainly be different for SO₂ and sulfate (ditto nitrogen species). Transfer velocity can be compared to measmts for wet. So you have the opportunity (and obligation) to compare modeled and measd conc and dep for wet; and conc for dry. And we need to know uncs on all quantities. The maps in Figs 6-7 and 6-8 are wholly insufficient.

In the scatter plot matrices I am concerned over choice of quantities to plot against each other; it seems of little sense or value to plot unrelated quantities such as nitrate dep vs sulfate conc. In general the slopes and uncertainties should be given, not just the correl coeff. Slope of dep vs conc in Fig 6-9 is transfer velocity. But need to do these by species. Cannot lump SO₂ and sulfate together; ditto N species. And should do wet and dry. For regulatory purposes there no need to waste a lot of time and effort doing this if well below stds, but from scientific perspective could learn a lot that is transferable from low conc and dep regions to high conc and dep regions and would lend confidence to application in those regions.

At the end of the day it is hard to tell what is the point of the scatter plot matrices. Do they argue on behalf of a particular approach to evaluating dep or do they argue against it. What is the required uncertainty?

Plots of ratio of, say, SO₂ conc sulfate conc, e.g. Fig 6-6, would convey much more information as fraction, $SO_2/(SO_2 + sulfate)$

In general, try to be at least semi-quantitative in characterizations, e.g., a term like "quickly" should be quantified, e.g. "hours to a day", "Differences tend to be small"; "larger differences".

In Section 6 I have expressed many concerns over the development of relation between design concentrations in source regions and deposition in receptor regions. Perhaps most fundamental is that the development of this relation is a major undertaking that in my judgment needs to be documented in scientific peer-reviewed literature rather than the results simply being presented here.

The development of the relation seems like a daunting task. Presumably hysplit trajectories every 6 hours, three levels. Determine conc in source regions that affect each Eco region. Transport material starting out at "design value" and do transformation and removal along the way for all source regions

that affect a given Eco region. These source regions would extend at least 1000 km along the back trajectory. Presumably not necessary to account for dispersion because the regions under consideration are so large. Calculate deposition in Eco region. All this needs to be spelled out in detail, presumably supported by appropriate references; too much detail required to present all that in this report. Transformation is a big deal, esp oxidation of SO₂ to sulfate, which requires knowledge of H₂O₂ and O₃ along the way. It does not seem that this report is the place for presentation of this research. It should be documented elsewhere and referred to here.

As noted in the specific comments, I have many scientific concerns over the approach. I end up with little confidence in the results presented.

Specific comments:

p. 1-1 – PM exerts cooling effect on climate through direct light scattering and modification of reflectivity and persistence of clouds. It is hard (for me) to make a connection between that and public welfare, but perhaps that should be explicitly noted.. I do not think reduction of solar irradiance is sufficient to have an effect on agriculture at least at current levels of aerosol optical depth, but I think that should be explicitly noted. There are exceptions: the recent smoke events (east coast) and prior year (california) had effects on health (not part of sec stds) but also visibility limiting aviation take-off and landing. This would seem to be within the remit of this report, but perhaps not within the remit of EPA, as not something that can be controlled by regulation, but it seems all of this requires explicit discussion.

I am surprised visibility is not included in this review. It would seem to be a major component of secondary effects of PM. It is not clear to me why visibility is not included.

p. 1-6, line 31 - the most recent date I see is 1988; has nothing happened since 1988? Report should state that. And should state what is the present standard.

p. 1-13, line 16 - What happened between 2018 and present?

p. 2-2, line 19 - I am not aware of any measurements showing 5% per hour in clear air.

p. 2-3, line 2 - and extents for in cloud oxidation; extent is more important than rate

p. 2-3, line 3 - relative to what? SO₂? that should be stated. But better estimates of lifetime would be maybe 1-2 days for SO₂, sink as dry dep; and about a week for sulfate; sink as wet dep. But this para should be informed, say, by the relations used in the evaluation of dry dep from concentrations later in this chapter.

p. 2-3, line 25 - again, a term like "quickly" should be quantified, eg "hours to a day", but again, such a statement should be informed by the dry dep modeling later in the chapter.

p. 2-4, line 19 - soil, vegetation surfaces, or

p. 2-6 – Figure 2-3 - It would be enormously informative to have a map like the Fig 2-3 for the several components of emission in the pie chart of Fig 2-2. My guess is that different source sectors have different geog distrib.

- p. 2-7, Figure 2-4 - This is enormously important tribute to the efforts of air quality regulation
- p. 2-9, Figure 2-6 – comment as in Fig. 2-3
- p. 2-10, Figure 2-7 – comment as in Fig. 2-4
- p. 2-11, Figure 2-9 – comment as in Fig. 2-3
- p. 2-12, Figure 2-10 - This figure is not useful; the "other" box must be subdivided into major subcategories
- p. 2-15 - I am surprised so sparse and unevenly distributed. Is this the 491 sites mentioned? Is this sufficient? Why so many in Wyoming; none in Iowa; the real question is whether the density of monitoring sites is sufficient to the needs. that should be addressed.
- p. 2-19, Figure 2-15 - can this fig be made similar layout to Figs 2.13 and 14. The thumb pins are distracting and obscure the information. Just enter the lat and lon coords and place them on a map. If the shading is of significance it should be explained.
- p. 2-20, Figure 2-16 - What is the significance of the yellow shading?
- p. 2-21, lines 2-3 - What is the accuracy with which dry dep can be determined from these measmts?
- p. 2-22, Figure 2-17 - comment as in Fig 2.17.
- p. 2-23 - What is definition of "Design Value"? The term is used throughout and I dont see a definition.
- p. 2-23, lines 8-9 – there a reason for that?
- p. 2-24 - looking for definition of design value; and why should design value differ from location to location? and what is a valid design value? I am guessing that a design value is a measurement according to some protocol. Seems like a misnomer. At minimum needs to be defined but perhaps a different name for the quantity.
- p. 2-26, Table 2-1 – wow. would be valuable to compare at same locations in present day measmts.
- p. 2-27 - what is a "valid design value"?
- p. 2-29 – Figure 2-26 - this runs the risk of being misleading, because of differing amts of pm2.5 at different locations. Can the circles be made such that the areas are proportional to pm2.5. We care less about the apportionment where the magnitude is small.
- p. 2-30, lines 3-5 - There is little EPA can do to control PM from wildfires, dust, sea spray, volcanoes. Perhaps the standard should exclude these components from the standard.
- p. 2-30, line 10 – change “while” to “whereas”
- p. 2-30, line 14 - Misuse of "significant"; better "substantial"; several instances in this para

p. 2-31 - Try for more consistency in figures. For example Fig 2-35 has scale at right; here underneath. Better at right

p. 2-32 - Fig 2-39 is much more information rich. If calculating dry dep then you have to calculate concs from which you can extract the 98th percentile information and plot it on a map and compare to the measmts.

p. 2-33, Figure 2-31 - Not very useful way of present data. Is there a better way,for example contours?

p. 2-33, Figure 2-32 - compare this fig to Fig 2-39; much more information rich.

p. 2-35 - Throughout: I am struggling to understand these figs because of lack of definition of "design value"

p. 2-36 - Again, aim for consistency in figures; Hard to read numbers on map; Color code is good; better low at bottom and high at top; No numbers on map but not sure they are needed.

p. 2-37, line 33 - That is impressive. Can we see a map of it?

p. 2-38, line 3 – change “particles” to “particulate.” What about NO_x?

p. 2-38, lines 9-11 - There should be (anti) correlation betw conc and dep velocity

p. 2-38, line 16 - How big is the bias?

p. 2-38, line 21 - It would seem that if constructing the 4 km or 12 km concentration data set for purpose of dry dep calcs, then that same data set could be used to infer concs for Air Quality.I dont see this done or referred to in sections on concentration standards. Is it done and I missed it or is it not done; and if not done, why not?

p. 2-38, line 25 – yes

p. 2-39 – Figure 2-37

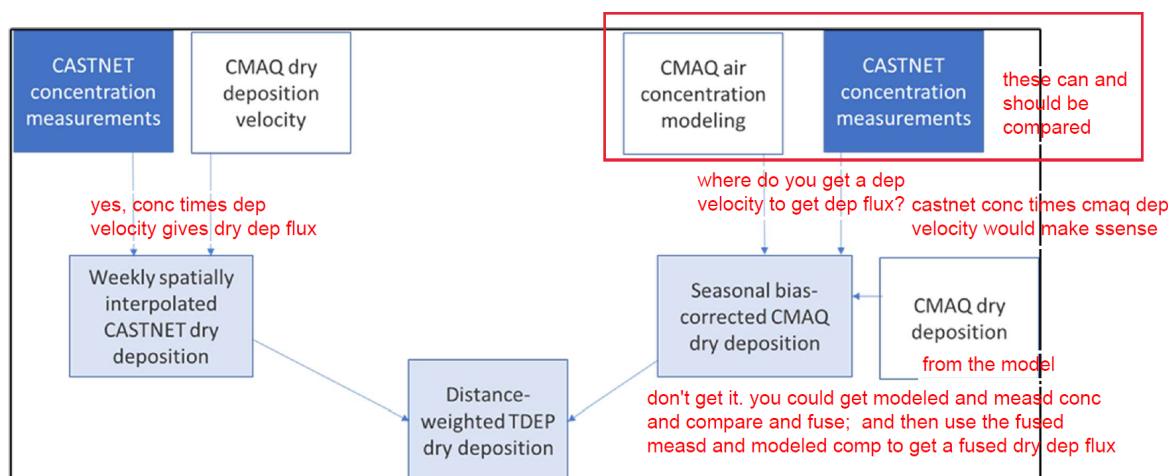


Figure 2-37. Data sources for estimating dry deposition. Dark blue indicates observations, white boxes indicate chemical transport modeling results, and light blue boxes are the results of model-measurement fusion. Note that the bias correction is not applied to ammonia, in part, because the existing method must be modified to account for its bidirectional flux.

are you ever comparing the model dry dep velocity with measmts of dry dep velocity by eddy correlation or gradient or the like?

p. 2-40, line 7 – “different models” – weak

p. 2-40, line 10 – much more robust

p. 2-40, line 27 – “latter compounds” – which? Be specific

p. 2-41, line 1 - If cmaq is used to calc dep velocities then the real evaluation of cmaq is comparison of measd and modeled concs, not dry deposition; yes? If so then should we be presented with those comparisons?

p. 2-41, lines 10-11 - yes, but you lose the impt spatial and temporal structure

p. 2-41, line 19 – “within 25% of the simulated values.” - That is impressive! at indiv locations and time or avgd over space and time? One would like to see maps comparing modeled and measd annual wet dep to assess this 25% statement.

p. 2-41, lines 22-23 – “bias” “55%” - which way? high or low? or is there some sort of map comparing? where do you assess the accuracy of the dry dep?

p. 2-42, lines 1-2 – “small” “larger” be specific; how small ; how much larger?

p. 2-42, line 24 – “quantify ecosystem effects” - As dry dep depends mainly on modeled concs, it seems imperative to have more discussion of the accuracy of dep velocities.

p. 2-43 - It would seem essential to show maps of wet dep and dry dep separately. I don't have a sense of the relative magnitudes. Wet dep is interpolated from measmts, whereas dry dep involves interpolated concs times dep velocities, which has inherently more uncertainty. If dry dep is small fraction, then the

large uncertainty in dry dep is of relatively little importance, but if dry dep is the large fraction, then that uncertainty looms large.

p. 2-43, Figure 2-38:

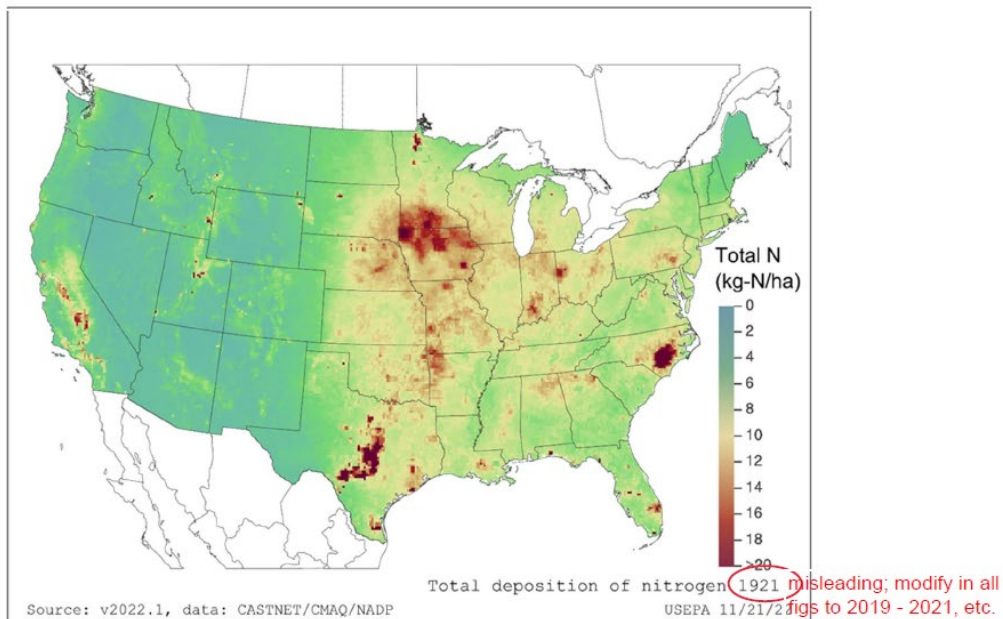


Figure 2-38. Three year average of the total deposition of nitrogen (kg N/ha) across the 2019-2021 period.

p. 2-45, Figure 2-40 - percent can be misleading; why not two maps, one showing reduced N dep, the other ox N dep. My guess is that much of the large red area may be small dep but that is just a guess. Or three maps: ox, red, total. In general have scale bars go from low at bottom to high at top.

p. 2-46 - Again, maps would help: maps of dep from international transport showing dep amount near borders, from lightning etc.

p. 2-47, Table 2-2:

Table 2-2. Change in total deposition by region between the 2000-2002 and 2019-2021 periods (U.S. EPA, 2022b): (a) total S deposition; (b) total, oxidized and reduced N deposition.

(a) Change in total S deposition		Very impressive		
Form of S Deposition	Region	2000-2002	2019-2021	% change
Total Deposition of Sulfur (kg S ha ⁻¹)	Mid-Atlantic	15.9	2.1	-87
	Midwest	11.2	2.2	-80
	North Central	3.5	1.5	-56
	Northeast	8.7	1.5	-83
	Pacific	1.0	0.6	-38
	Rocky Mountain	1.0	0.6	-46
	South Central	5.4	2.8	-49
	Southeast	10.3	2.6	-74
(b) Change in total, oxidized and reduced N deposition				
Form of N Deposition	Region	2000-2002	2019-2021	% change
Total Deposition of Nitrogen (kg N ha ⁻¹)	Mid-Atlantic	13.4	8.5	-36
	Midwest	12.2	9.8	-20
	North Central	8.5	9.5	+11
	Northeast	10.4	6.2	-40
	Pacific	3.8	3.1	-18
	Rocky Mountain	3.0	3.1	+3
	South Central	7.8	9.0	+16
	Southeast	10.8	8.4	-23
Total Deposition of Oxidized Nitrogen (kg N ha ⁻¹)	Mid-Atlantic	10.3	4.0	-62
	Midwest	8.0	3.6	-54
	North Central	4.1	2.6	-37
	Northeast	7.7	2.9	-62
	Pacific	2.4	1.4	-42
	Rocky Mountain	1.9	1.3	-35
	South Central	5.0	3.1	-39
	Southeast	7.7	3.4	-56
Total Deposition of Reduced Nitrogen (kg N ha ⁻¹)	Mid-Atlantic	3.0	4.6	+51
	Midwest	4.3	6.2	+45
	North Central	4.4	6.9	+56
	Northeast	2.7	3.3	+22
	Pacific	1.4	1.7	+22
	Rocky Mountain	1.1	1.8	+72
	South Central	2.8	6.0	+111
	Southeast	3.1	5.0	+63
Why is this going up?				

p. 2-49, Figure 2-42 - why the hot spots of deposition where there doesnt seem to be corresponding hot spots in map of HNO3 conc? in fig 2-41

p. 2-50, line 12 – “projecting future trends” - Is this necessary in this report which deals with standards setting?

p. 2-51, Figure 2-43 - again percent can be misleading. I think much better to show amounts in each time period.

p. 2-52, Figure 2-44 - are such projections part of the remit of this study, which focuses on science relevant to secondary stds

p. 3-4, lines 25-28 – “transference ratios,” - That is interesting approach. As expressed later I am concerned over using transference ratio for multiple compounds; transference ratio would seem to be applicable only to a single compound. Also separately for wet and dry dep. At least from science perspective. Perhaps justified from an engineering perspective, provided the consequences of lumping multiple compounds and wet and dry dep are suitably assessed.

p. 3-8, line 5 – “neither more nor less stringent than necessary” - With respect to stringency: Given inevitable uncertainties, how does the standard take these into account? Do you make the standard more stringent to set the max allowable conc equal to the best estimate minus the unc; or less stringent to make the max allowable conc equal to the best estimate plus the unc? Or just ignore the unc and make the max allowable conc equal to the best estimate value? I think that this needs to be explicitly stated.

p. 5-5, line 5 – “exposures to 30 ppb PAN on alternating days” - I recall field studies showing effects of PAN on leafy crops such as spinach in Santa Clara Calif as early as the 50's or 60s. Are any of those studies relevant here?

p. 5-6, line 9 - At least mention reduction of solar irradiance and changes between direct and indirect irradiance

p. 5-14, line 1 – change “were” to “are

p. 5-16, line 28 - Just thinking: Critical load is in units of Eq m⁻² yr⁻¹, but ANC is in units Eq L⁻¹ or Eq m⁻³. So somehow the relation between critical load and ANC needs to take into account the depth of the water body (to relate area to volume) but also something to account for time, something like the rate at which ANC is added to the water body, which would seem to involve leaching of minerals or the like. At very least it would seem that a para describing the relation is required.

p. 5-18, line 5 - steady state concept implies some sort of rate of production of ANC; see comment above for p. 5-16

p. 5-19, line 14 - Suggest avoid first person plural. Maybe better "EPA compared" or even better passive voice " waterbody estimates are compared to critical loads." or better, "This analysis compares"

p. 5-20, line 12 – “3.125 meq S/m²-yr” I am reminded here of LaHerrere's law: any author giving more than 3 significant digits shows that he is incompetent in assessing accuracy and in probability. Usually when more than 3 decimal digits are used, it is likely that the first digit is wrong.

p. 5-20, line 16 - Does deposition rate include the catchment area of the water body?

p. 5-21, Figure 5-6 - How do you get from ANC to critical load? Perhaps I missed it. See comment for p. 5-16

p. 5-32, line 1 - avoid first person plural throughout

p. 6-2, lines 27-29 - I disagree strenuously with that statement. For a gaseous species the deposition rate (for a given amount of precep) depends on the solubility (Henry's law or Henry's law extended to account for acid or base dissociation). As an extreme, the transfer of N₂ to cloudwater and into precip is much less than that of HNO₃. NO and NO₂ are somewhere inbetween. NO and NO₂ are not very soluble in cloudwater. Oxidation to the very soluble HNO₃ is mainly in gas phase. SO₂ is likewise not very soluble, even taking into account dissociation equilibria, but is rapidly oxidized in cloudwater if there is sufficient H₂O₂; ozone oxidation is slower.

p. 6-2, line 31 - "settle" - Not a good verb; the dry dep process involves chem rxn with surface material, leaves, etc.

p. 6-2, line 33 - "via" - Not so much "via" ; that scavenging is in fact the wet dep process

p. 6-3, line 1 - be specific: sulfur and nitrogen oxides can be emitted by tall stacks; tailpipes might be considered ground level. Both are distributed throughout the atmos boundary layer on time scale of a day; SO₂ and NO_x can be transported for hundreds of km, during which time there can be (and is) substantial oxidation to sulfate and HNO₃, with resultant wet dep.

p. 6-3, line 2 - "Further" - try to be a little more specific here: hundreds of km

p. 6-3, line 10 - "Transference Ratio" - It seems essential to present transference ratio in this document, not just refer to another document. Transference Ratio would be quotient (not ratio!) of dep flux to air conc and a useful unit would be g m⁻² yr⁻¹/(g m⁻³) or m yr⁻¹ or cm s⁻¹, so even better to call it a transfer velocity, not transference ratio. And yes it seems essential to present maps of annual transfer velocity so we could see the spatial variability; time series of transfer velocity at a few representative locations. Show at fairly high spatial resolution so we could see the effects of larger spatial averaging; this would be important if concs are spatially inhomogenous. It would seem that transfer velocity for a gaseous species is the same as the deposition velocity. So extension to wet dep permits comparison. So show for both wet and dry dep. And it seems almost certain that these transfer velocities will be different for each chemical species. I briefly looked at the integrated science assessment referenced here. It states "These ratios are expressed in units of distance/time (as a velocity)," rather as I deduced above. It does not seem to give values for these quantities. It does explicitly acknowledge that the transference ratio depends on the mix of substances, e.g., SO₂ and sulfate; that in itself would seem to be a fatal flaw in the concept. Figure 2-11 of that document gives plots of TR evaluated by one model vs another (units not specified). I infer that the variability of TR has to do with the mix of species. I would have many criticisms of that approach. It refers to journal articles by Koo and Sickles. I expect I would have similar concerns there too. But for the present review it seems incumbent on the authors to justify why this approach (greatly flawed in my judgment) can be relied on for the purpose of standard setting. At minimum this would require assessment of uncertainties resulting from application of transference ratio concept to mix of species with very different dep velocities. I think it is essential to present maps of transference ratio as a function of location, if, as I understand it, TR depends on mix of species, which in turn depends on distance from sources among other factors.

p. 6-3, line 13 - "10,000 km²." - Distance is km; area is km²; Take your pick. Might depend on the area over which transference ratio is calculated.

p.6-3, line 16 - "proximity to sources" - why is that imp? composition is key; vert distrib is key.

p. 6-4, lines 13-14 - show individually, not just the sum

p. 6-4, line 17 - how much error does this introduce?

p. 6-4, lines 27-28 - not useful; what we need is comparison of modeled and measd dep; also a map showing wet and dry of S and N individually, to support the statement here

p. 6-4, line 29 - what are the statistics on these numbers? These are very impt conclusions. What is the evidence, and what is the spread in ratios, geographically etc. certainly need to give uncertainties on these numbers; based on fig 6-3 they would seem to be huge

p. 6-4, Table 6-1 - better in an appendix. this table would be greatly enhanced by small box whisker plots similar to Fig 6-3 for each location.

p. 6-6, Figure 6-2 - Useless to show the map of stations. Need to see the data as a fn of location

p. 6-6, Figure 6-3 - not very useful

p. 6-7, line 5 - YES!!

p. 6-7, line 10 - Well, the slope itself, not just the unc in the slope

p. 6-7, line 19-20 – “Figure 6-3.” - No. Fig 6-3 shows wet and dry dep. Figure 6-5?

p. 6-8, lines 1-2 - This an impt result, but the imptce is not the correlation coeff but the slope, kg ha-1 yr-1 / ($\mu\text{g m}^{-3}$) or meters per year. That is the quantity I would like to see. I guess annual average is ok if the proposed standard is annual deposition.

p. 6-8, line 7 – “Figure 6-5” – Figure 6-6?

p. 6-8 lines 10-11 - yes, but show. My guess is that the fraction of $\text{SO}_2/(\text{SO}_2 + \text{sulfate})$ is very strongly spatially dependent. Why not show for each station. And perhaps dependent on time given that the data stretch from 1998 to 2018.

p. 6-9, Figure 6-4:

suggest give slope and uncertainty. For deposition vs concentration it would seem that a useful unit for the slope would be m yr⁻¹

I dont see any value of plotting S and N on same scatterplot matrix.

Is this poster child of good correl simply a consequence of N being << S, so it is essentially a plot of S vs S

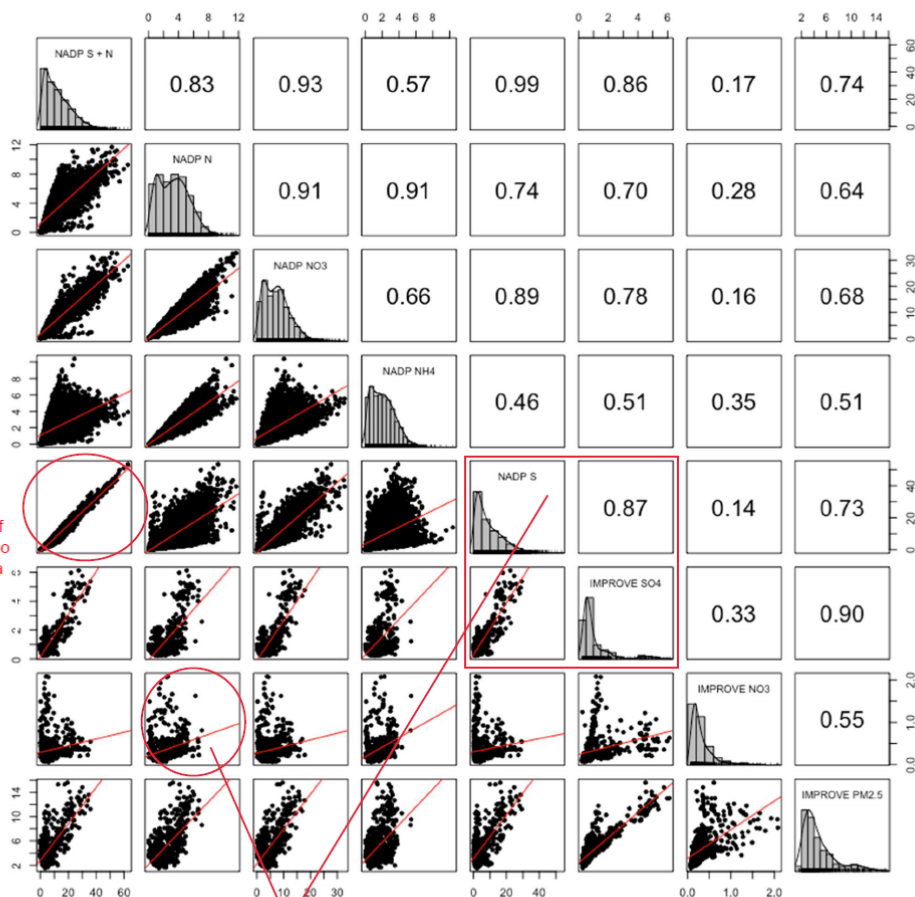


Figure 6-4. Scatter plot matrix of annual average wet deposition measurements from NADP/NTN (5 pollutants, units: kg/ha-yr) versus annual average concentrations from IMPROVE (3 pollutants, units: µg/m³) for 27 Class 1 areas from 1988-2018. A histogram of each deposition or concentration variable is shown in a diagonal running from the top left to lower right. Below that diagonal are scatter plots for each pair of variables. Above that diagonal are the correlations between pairs of variables. (Note for this plot and all subsequent matrix plots: the x- and y- axes scales are shown on the left and right sides of the plot for rows, and at the top and bottom of the plot for columns.)

In contrast this shows essentially no skill

This seems to be an important correl; gives a lot of confidence

May 2023

6-9

Draft – Do Not Quote or Cite

It seems you are showing a lot of irrelevant stuff here instead of focusing on the important features

p. 6-10, Figure 6-5:

I dont see any value of
plotting S and N on same
scatterplot matrix.

Is this poster
child of good
correl simply a
consequence of
N being << S, so
it is essentially a
plot of S vs S

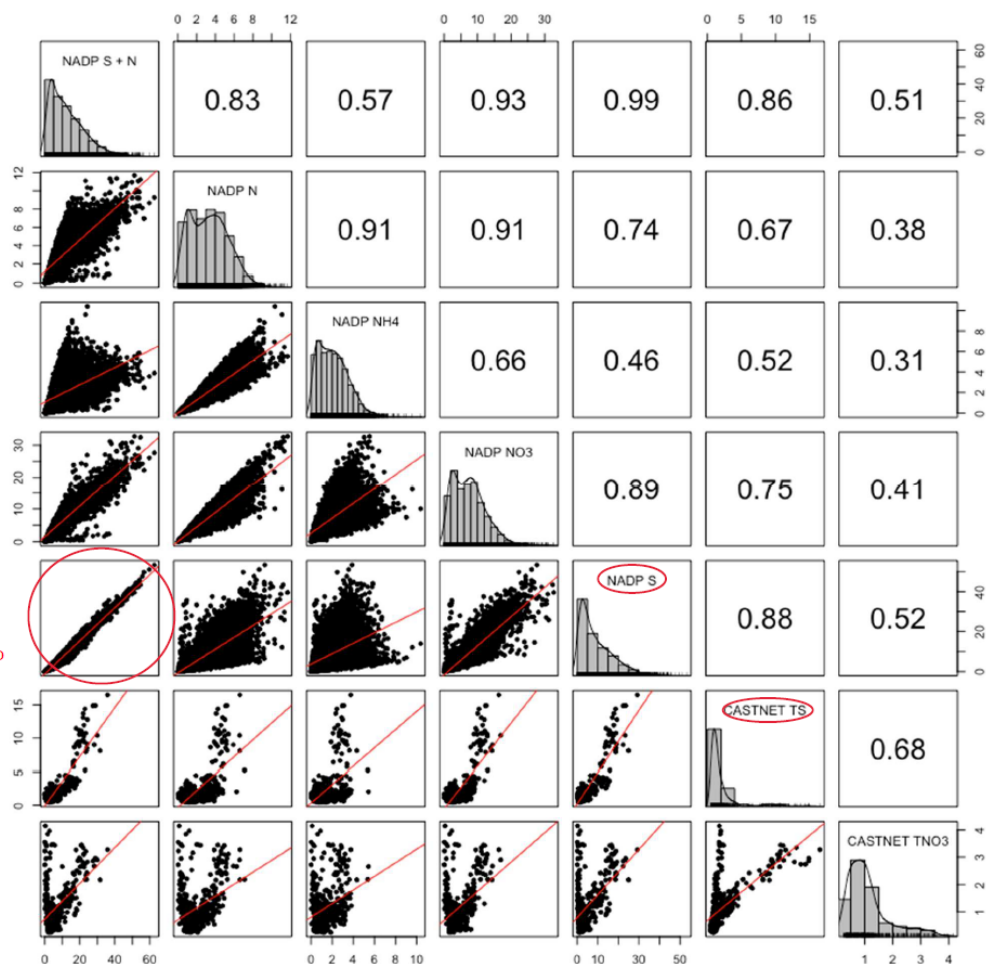


Figure 6-5. Scatter plot matrix of annual average wet deposition measurements from NADP/NTN (5 pollutants, units: kg/ha-yr) versus annual average concentrations from CASTNET (2 pollutants, units: µg/m³) for 27 Class 1 areas from 1988-2018. A histogram of each deposition or concentration variable is shown in a diagonal running from the top left to lower right. Below that diagonal are scatter plots for each pair of variables. Above that diagonal are the correlations between pairs of variables.

could the low correl betw NADP total S and castnet total S be due
to presence of SO2; what if plot NADP total S vs castnet sulfate?
What does this low correl tell you about accuracy of inferring
sulfate wet dep from modeled total S conc?

p. 6-11, Figure 6-6:

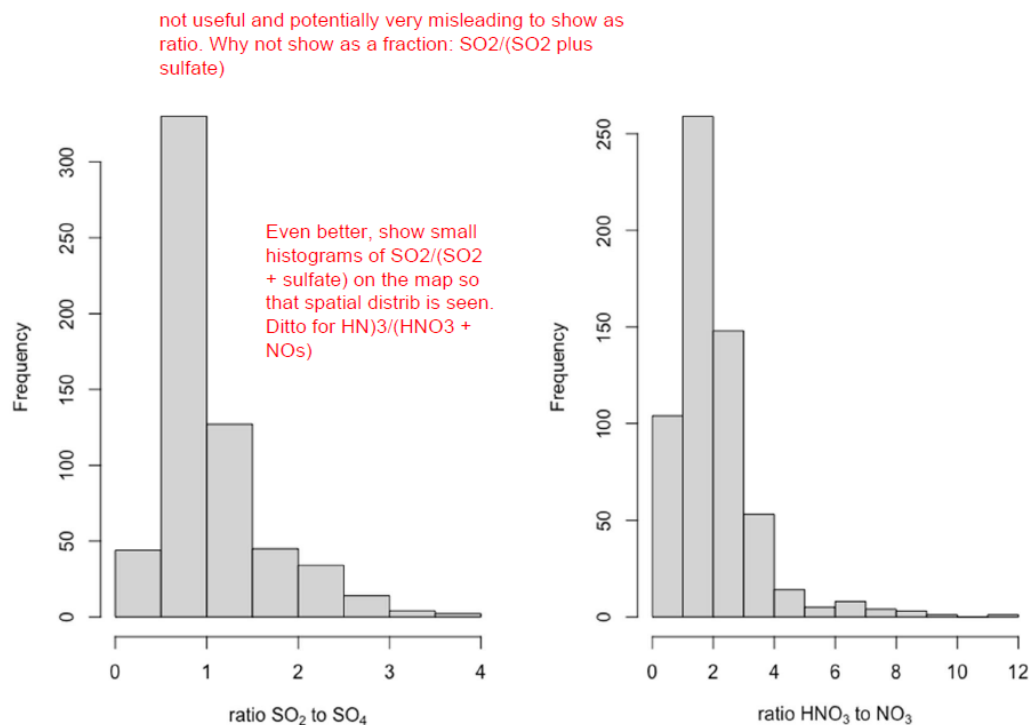


Figure 6-6. Histograms of the ratios of the gas phase SO_2 to particle SO_4^{2-} (left) and the gas phase HNO_3 to particle NO_3^- (right) in CASTNET data. Each ratio is calculated as the annual average concentration (2000-2019), converted to moles of N or moles of S, for the 27 sites listed in Table 6-1.

p. 6-11, line 10 - “not” - why not? esp if used as an argument

p. 6-11 line 18 – p. 6-12, line 2 - should this para precede the previous one? This states the observations. The prev para tries to explain the obs. Yes?

p. 6-12, line 4 - Yes, rarely measd; all the more reason to compare model to meas when measd.

pp. 6-13 – 6-14 – Figures 6-7 and 6-8 - There are two important steps in the CMAQ modeling: modeling the concentration as a function of location and time, and then modeling the deposition as a function of location and time. The quotient is the transfer velocity. So all should be shown. To be sure we don't have the measurement of dry deposition flux, so we have to rely on the model, but we need to have some measure of the confidence in the model. If the modeled dry deposition is well below the standard deviation, then we don't much care how accurate the model is, but if it is comparable to the standard deviation, then it becomes important and needs to be assessed. The ratio should have units (I suggest m yr^{-1}) and presented for wet and dry, and it should certainly be different for SO_2 and sulfate (ditto nitrogen species). Transfer velocity can be compared to measurements for wet. So you have the opportunity (and obligation) to compare modeled and measured concentration and deposition for wet; and concentration for dry. And we need to know uncertainties on all quantities. The maps in Figs 6-7 and 6-8 are wholly insufficient.

p. 6-13, Figure 6-7:

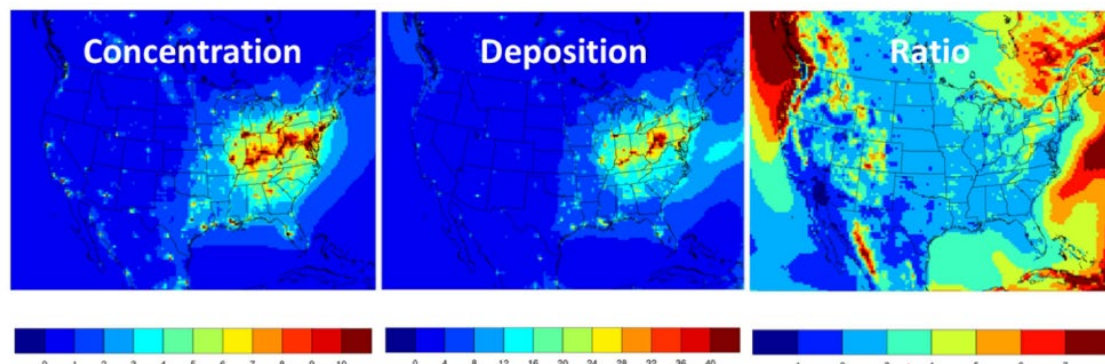


Figure 6-7. Annual average concentration ($\mu\text{g}/\text{m}^3$), deposition ($\text{kg}/\text{ha}\cdot\text{yr}$), and the deposition/concentration ratio for oxidized sulfur compounds, as estimated using a 21-year (1990-2010) CMAQ simulation.

what means? SO_2 + sulfate? stoichiometry question if includes both in mass. Better moles.

units? must be specified. $\text{kg ha}^{-1}\text{yr}^{-1}/(\mu\text{g m}^{-3})$? or better m yr^{-1}

p. 6-15, line 4 – better, passive voice

p. 6-15, line 9 – where shown?

p. 6-15, line 15-16 – avoid first person plural

p. 6-15, line 25 - show separately by species instead of total; it will strengthen your conclusions.

p. 6-16, Figure 2-9 - Similar comments as on earlier scatter plots; show slopes and uncs. Slope of dep vs conc is transfer velocity. But need to do these by species. Cannot lump SO_2 and sulfate together; ditto N species. And should do wet and dry. Again (for regulatory purposes) no need to waste a lot of time and effort doing this if well below stds, but from scientific perspective could learn a lot that is transferable from low conc and dep regions to high conc and dep regions and would lend confidence to application in those regions. should also examine as a function of time and location to see if any systematic differences.

p. 6-18, Figure 6-10:

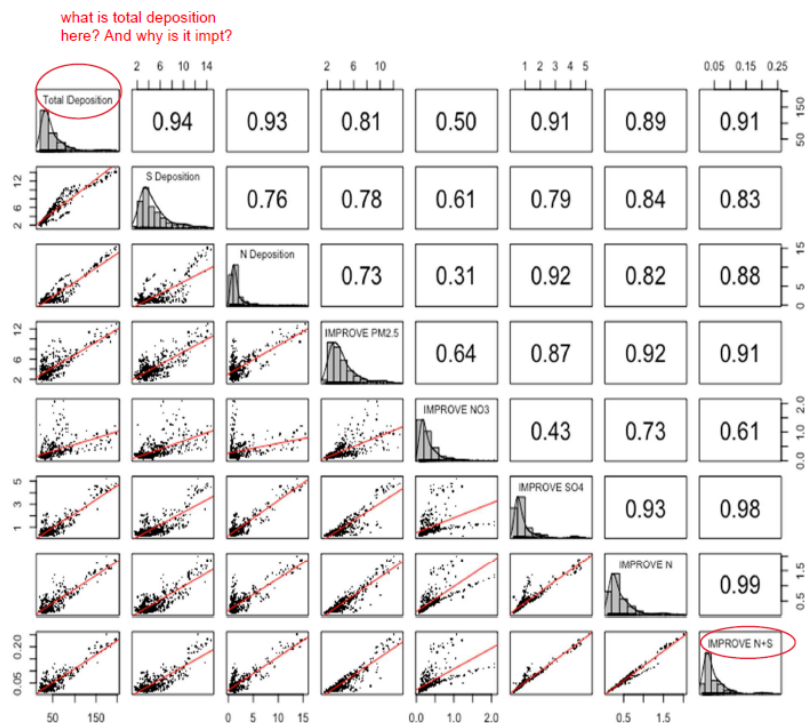


Figure 6-10. Scatter plot matrix of annual average TDEP deposition (3 pollutants, units: kg/ha-yr) versus annual average IMPROVE concentrations (5 pollutants, units: $\mu\text{g}/\text{m}^3$) for 27 Class 1 areas with collocated IMPROVE and NADP/NTN from 1988-2018. A histogram of each deposition or concentration variable is shown in a diagonal running from the top left to lower right. Below that diagonal are scatter plots for each pair of variables. Above that diagonal are the correlations between pairs of variables.

Is this wet and dry?
should be shown
separately. And by
species.

p. 6-20, line 14 - Avoid first person plural. Inherently ambiguous whose understanding? The authors; EPA; the scientific community?

p. 6-20, lines 30-31 - Try to be quantitative in assessment. With what accuracy. Factor of 2? With what bias? Do these findings support the approach of using air conc to limit deposition to protect sensitive watersheds, or to back calculate the maximum allowable air conc that would be protective?

p. 6-22, line 7 - Still don't know what is definition of design value

p. 6-23 - This seems like a daunting task. Presumably hysplit trajectories every 6 hours, three levels. Determine conc in source regions that affect each Eco region. Transport material starting out at "design value" and do transformation and removal along the way for all source regions that affect a given Eco region. These source regions would extend at least 1000 km along the back trajectory. Presumably not necessary to account for dispersion because the regions under consideration are so large. Calculate deposition in Eco region. All this needs to be spelled out in detail, presumably supported by appropriate references; too much detail required to present all that in this report. Transformation is a big deal, esp oxidation of SO_2 to sulfate, which requires knowledge of H_2O_2 and O_3 along the way. It does not seem

that this report is the place for presentation of this research. It should be documented elsewhere and referred to here.

p. 6-24 - Again the nature of the calc is not well spelled out. Are you suggesting that each upwind location is at its maximum design value conc for the entire period during which the calc is made? That would lead to enormous over prediction of deposition at receptor location. Or are you assuming statistics on the source region concs? It seems enormously daunting to calc cumulative dep by running this model for three years t 6 hr intervals for each contributng source region. In the absence of suitable scientific documentation of the model and its application I have little confidence in the results.

p. 6-25 - For deposition there are multiple sites of influence and deposition can (and does) come from multiple upwind sites along the back trajectory. Does this analysis take that into account?

p. 6-26, line 1 – “Figure 6-16...” - Suggest new para; this is a different subject

p. 6-27 - might be more useful to present the inverse quantity that represents the fractional decrease of SO₂ between source and receptor.

p. 6-30 - It would seem that doing these calcs for N deposition is even more daunting than for S dep, given the role of oxidation of NO₂ to HNO₃

p. 6-32 - Hard to see the basis of expected correl. PM_{2.5} consists of sulfate, nitrate, organics, the latter can comprise substantial fraction. See for example Zhang Zhang, Q. Ubiquity and dominance of oxygenated species in organic aerosols in anthropogenically-influenced Northern Hemisphere midlatitudes. Geophysical research letters, 34(13). So hard to make the connection betw PM_{2.5} and sulfate or S+ N dep. Also N dep includes HNO₃ not included in PM_{2.5}

p. 6-34 - this is astonishingly strong correl, but as mentioned, PM contains much more than N, and N dep also from HNO₃; can this be coincidence?

p. 6-38, lines 19-20 – “close” “further away” - Again try to be at least semi-quantitative; close to (eg < 300 km); further away (>300 km) or something like that

p. 5A-17, line 27 - need time and area

p. 5A-24 - All these tables seem to make the eye glaze over. Perhaps they are necessary, but can the information also be presented in maps?

Dr. Kathleen Weathers

Chapter 3 – Current Standards and Approach

To what extent does the Panel find that the background information in sections 3.31 and 3.2 to be clearly presented and provide useful context for the review?

The background information is mostly well- and clearly- presented and informative. However, in the first sentence (lines 3 and 4) it would be helpful to be reminded of why the difference between consideration of (full complement of?) *welfare effects* for oxides of S and N and the (subcategory) of *ecological* effects for PM. While there is a little more information at the end of section 3.1, the why is not clear (see additional comment below)

It was especially useful to be reminded in the first paragraph in section 3.1 that the early secondary standards were set based on vegetation (+crops for NO_x) effects, despite other (unquantifiable but associated) welfare effects.

It would be helpful to add a sentence or two about the logic and/or history for parsing welfare effects for PM secondary standards, and the focus, here, only on ecological effects. I suggest also adding a phrase with the background for the statement on line 15 “prior reviews have generally concluded...”.

I appreciated the description of the prior review of deposition effects. It is succinct and reasonably clear for what was a complicated analysis. A paragraph that reveals some of the biggest model and data uncertainty stumbling blocks is important to add.

What are the Panel’s views on the general approach described in section 3.3 for considering the welfare effects evidence and the risk assessment information for review of the adequacy of the current NO₂, SO₂ and PM secondary standards?

I find the approach relevant and logical. There are also at least two things that I think are important advances in this review, which are that deposition-related effects are the focus, and that PM has been added, despite the fact that there are complications with the non S- or N-composition of PM.

It’s not entirely clear what is meant on line 13 by “both evidence- and exposure/risk-based considerations. I suggest adding at least a phrase that outlines what is meant. The same is true for “quantitative information.”

Figure 3-1: I cannot discern whether the different box shapes have meaning. If they do, please identify in the legend. If they don’t, I suggest standardizing. (I find them distracting--as I try to discern meaning for them). Also, the 4th bullet in the “Evidenced-Based Considerations box redoubles my interest in seeing a paragraph in earlier sections describing a little more specifically the uncertainty issues identified previously (see comment above).

Figure 3-2: Quantitative information/analyses still reads as too vague to me, as does “model-estimated spatial relationships.”

Page 3-10, lines 10-14 and the next paragraph rightly identifies an important and extremely knotty historical/legacy/lag issue.

Page 3-11, line 6: The parenthetical “steady-state water quality modeling” should be pulled out of the parens. Also, consider adding an additional sentence that says what is meant by steady state, and maybe point to a discussion somewhere else in the document about the limitations and/or uncertainties (and benefits) of such a model.

Similarly, lines 9-12, identify what “air quality modeling” and how it relates to “tracks” (i.e., Hysplit tracks?)

I also applaud the range of potential policy options that are proposed here (and laid out in chapter 7).

Chapter 7 – Review of the Standards

What are the Panel views on the discussion of key considerations in sections 7.1 and 7.2 and preliminary conclusions?

Included in the beginning part of this chapter should be a sentence or two that acknowledges that while a deposition standard for welfare effects was not for the last review, and is not now being considered, CASAC strongly recommends that EPA do so in the future. Most welfare effects are driven by the actual delivery of S, N, and particulates rather than exposure.

This chapter describes well the various policy options relevant to welfare effects. Particularly useful was the description of policy options and associated Table 7.1.

Section 7.1 was informative, but I was confused by the set of focal questions to be answered (the bolded/bulleted questions). While I agree that they are relevant and important, the answers to them were not clearly highlighted in the text below. A particularly important omission (or burying), to my read, was a discussion with regard to “to what extent have important uncertainties...”

Page 7-2: Lines 3 and 6: is there a difference between “newly available” and “currently available” information? If so, what is it? If not, I suggest you use the same word.

In the first paragraph in 7.1.1 I’m still not sure whether there is any newly or currently available information and if so, what it is? Please clarify. (And now I see the answer, I believe, in the 2nd paragraph—the answers to the bold/bulleted questions could still be made clearer and more direct.)

Please also clarify whether laboratory studies are the same as experimental studies and observational studies are synonymous with field studies --and make sure to use the words consistently throughout.

I appreciated the clear beginning to the first paragraph in section 7.1.2.

Page 7-4, Line 35: “While new uncertainties...” this sentence is quite vague.

Section 7.1.3: does this section refer to both PM10 and PM2.5? Clarify. Also, the last paragraph in this section should be rewritten for clarity.

Section 7.2 was also informative and mostly clear, however the first paragraph was a bit awkward (vis a vis commentary on the organization of the discussion).

Section 7.2.1: What is meant by “quantitative analyses/aspects” should be explicated (in earlier chapters if not again here).

What are the CASAC views regarding the areas for additional research identified in Chapters 3 and 4? Are there additional areas that should be highlighted?

The paragraph introducing Section 7.4 should be more explicit about the need for expanded in-situ monitoring and measurement, atmospheric chemistry, atmospheric deposition and exposure research as well as model development, comparison and validation research. I suggest that the bulleted list begin with broad categories of research needs and then underneath include specifics (e.g., the spatial gaps for monitoring/measurement associated with the pollutants that need to be included.)

Also included in the additional research needs should be (1) development and adoption of new models and modeling tools and approaches, such as the application of knowledge-guided machine learning, and, importantly (2) assessments of the adequacy and/or comparison of models (e.g., steady state with dynamic biogeochemical models).