The EPA Administrator, Lisa P. Jackson, signed the following notice on 07/12/2011, and EPA is submitting it for publication in the *Federal Register* (FR). While we have taken steps to ensure the accuracy of this Internet version of the rule, it is not the official version of the rule for purposes of compliance. Please refer to the official version in a forthcoming FR publication, which will appear on the Government Printing Office's FDSys website (http://fdsys.gpo.gov/fdsys/search/home.action) and on Regulations.gov (http://www.regulations.gov) in Docket No. **EPA-HQ-OAR-2007-1145**. Once the official version of this document is published in the FR, this version will be removed from the Internet and replaced with a link to the official version.

6560-50-P

ENVIRONMENTAL PROTECTION AGENCY

40 CFR Part 50

[EPA-HQ-OAR-2007-1145]

RIN: 2060-AO72

Secondary National Ambient Air Quality Standards for Oxides of Nitrogen and Sulfur

AGENCY: Environmental Protection Agency (EPA).

ACTION: Proposed rule.

SUMMARY: This proposed rule is being issued as required by a consent decree governing the schedule for completion of this review of the air quality criteria and the secondary national ambient air quality standards (NAAQS) for oxides of nitrogen and oxides of sulfur. Based on its review, EPA proposes to retain the current nitrogen dioxide (NO_2) and sulfur dioxide (SO_2) secondary standards to provide requisite protection for the direct effects on vegetation resulting from exposure to gaseous oxides of nitrogen and sulfur in the ambient air. Additionally, with regard to protection from the deposition of oxides of nitrogen and sulfur to sensitive aquatic and terrestrial ecosystems, including acidification and nutrient enrichment effects, EPA is proposing to add secondary standards identical to the NO_2 and SO_2 primary 1-hour standards and not set a new multi-pollutant secondary standard in this review. The proposed 1-hour secondary NO_2 standard would be set at a level of 100 ppb and the proposed 1-hour secondary SO_2 standard would be set at 75 ppb. In addition, EPA has decided to undertake a field pilot program to gather

and analyze additional relevant data so as to enhance the Agency's understanding of the degree of protectiveness that a new multi-pollutant approach, defined in terms of an aquatic acidification index (AAI), would afford and to support development of an appropriate monitoring network for such a standard. The EPA solicits comment on the framework of such a standard and on the design of the field pilot program. The EPA will sign a notice of final rulemaking for this review no later than March 20, 2012.

DATES: Written comments on this proposed rule must be received by **[INSERT DATE 60 DAYS AFTER DATE OF PUBLICATION IN THE FEDERAL REGISTER]**.

Public Hearings: The EPA intends to hold a public hearing around the end of August to early September and will announce in a separate Federal Register notice the date, time, and address of the public hearing on this proposed rule.

ADDRESSES: Submit your comments, identified by Docket ID No. EPA-HQ-OAR-2007-1145, by one of the following methods:

- www.regulations.gov: Follow the on-line instructions for submitting comments.
- Email: a-and-r-Docket@epa.gov.
- Fax: 202-566-1741.
- Mail: Docket No. EPA-HQ-OAR-2007-1145, Environmental Protection Agency, Mail code 6102T, 1200 Pennsylvania Ave., NW, Washington, DC 20460. Please include a total of two copies.
- Hand Delivery: Docket No. EPA-HQ-OAR-2007-1145, Environmental Protection Agency, EPA West, Room 3334, 1301 Constitution Ave., NW, Washington, DC. Such

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deliveries are only accepted during the Docket's normal hours of operation, and special arrangements should be made for deliveries of boxed information.

Instructions: Direct your comments to Docket ID No. EPA-HQ-OAR-2007-1145. The EPA's policy is that all comments received will be included in the public docket without change and may be made available online at www.regulations.gov, including any personal information provided, unless the comment includes information claimed to be Confidential Business Information (CBI) or other information whose disclosure is restricted by statute. Do not submit information that you consider to be CBI or otherwise protected through www.regulations.gov or email. The www.regulations.gov website is an "anonymous access" system, which means EPA will not know your identity or contact information unless you provide it in the body of your comment. If you send an email comment directly to EPA without going through www.regulations.gov, your email address will be automatically captured and included as part of the comment that is placed in the public docket and made available on the Internet. If you submit an electronic comment, EPA recommends that you include your name and other contact information in the body of your comment and with any disk or CD-ROM you submit. If EPA cannot read your comment due to technical difficulties and cannot contact you for clarification, EPA may not be able to consider your comment. Electronic files should avoid the use of special characters, any form of encryption, and be free of any defects or viruses. For additional information about EPA's public docket, visit the EPA Docket Center homepage at http://www.epa.gov/epahome/dockets.htm.

Docket: All documents in the docket are listed in the www.regulations.gov index. Although listed in the index, some information is not publicly available, e.g., CBI or other information whose disclosure is restricted by statute. Certain other material, such as copyrighted

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material, will be publicly available only in hard copy. Publicly available docket materials are available either electronically in www.regulations.gov or in hard copy at the Air and Radiation Docket and Information Center, EPA/DC, EPA West, Room 3334, 1301 Constitution Ave., NW, Washington, DC. The Public Reading Room is open from 8:30 a.m. to 4:30 p.m., Monday through Friday, excluding legal holidays. The telephone number for the Public Reading Room is (202) 566-1744 and the telephone number for the Air and Radiation Docket and Information Center is (202) 566-1742.

FOR FURTHER INFORMATION CONTACT: Dr. Richard Scheffe, Office of Air Quality Planning and Standards, U.S. Environmental Protection Agency, Mail code C304-02, Research Triangle Park, NC 27711; telephone: 919-541-4650; fax: 919-541-2357; email: scheffe.rich@epa.gov.

SUPPLEMENTARY INFORMATION:

General Information

What Should I Consider as I Prepare My Comments for EPA?

1. *Submitting CBI.* Do not submit this information to EPA through www.regulations.gov or email. Clearly mark the part or all of the information that you claim to be CBI. For CBI information in a disk or CD ROM that you mail to EPA, mark the outside of the disk or CD ROM as CBI and then identify electronically within the disk or CD ROM the specific information that is claimed as CBI. In addition to one complete version of the comment that includes information claimed as CBI, a copy of the comment that does not contain the information claimed as CBI must be submitted for inclusion in the public docket. Information so marked will not be disclosed except in accordance with procedures set forth in 40 CFR part 2.

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2. *Tips for Preparing Your Comments*. When submitting comments, remember to:

- Identify the rulemaking by docket number and other identifying information (subject heading, Federal Register date and page number).
- Follow directions – The Agency may ask you to respond to specific questions or organize comments by referencing a Code of Federal Regulations (CFR) part or section number.
- Explain why you agree or disagree, suggest alternatives, and substitute language for your requested changes.
- Describe any assumptions and provide any technical information and/or data that you used.
- If you estimate potential costs or burdens, explain how you arrived at your estimate in sufficient detail to allow for it to be reproduced.
- Provide specific examples to illustrate your concerns, and suggest alternatives.
- Explain your views as clearly as possible.
- Make sure to submit your comments by the comment period deadline identified.

Availability of Related Information

A number of documents relevant to this rulemaking are available on EPA web sites. The Integrated Science Assessment for Oxides of Nitrogen and Sulfur - Ecological Criteria: Final Report (ISA) is available on EPAs National Center for Environmental Assessment web site. To obtain this document, go to http://www.epa.gov/ncea, and click on Air Quality then click on Oxides of Nitrogen and Sulfur. The Policy Assessment (PA), Risk and Exposure Assessment (REA), and other related technical documents are available on EPA's Office of Air Quality Planning and Standards (OAQPS) Technology Transfer Network (TTN) web site. The PA is

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available at http://www.epa.gov/ttn/naaqs/standards/no2so2sec/cr_pa.html, and the exposure and risk assessments and other related technical documents are available at http://www.epa.gov/ttn/naaqs/standards/no2so2sec/cr_rea.html. These and other related documents are also available for inspection and copying in the EPA docket identified above.

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I. Background

A. Legislative Requirements

Two sections of the Clean Air Act (CAA) govern the establishment and revision of the NAAQS. Section 108 (42 U.S.C. section 7408) directs the Administrator to identify and list certain air pollutants and then to issue air quality criteria for those pollutants. The Administrator is to list those air pollutants that in her "judgment, cause or contribute to air pollution which may reasonably be anticipated to endanger public health or welfare;" "the presence of which in the ambient air results from numerous or diverse mobile or stationary sources;" and "for which . . . [the Administrator] plans to issue air quality criteria…" Air quality criteria are intended to "accurately reflect the latest scientific knowledge useful in indicating the kind and extent of all identifiable effects on public health or welfare which may be expected from the presence of [a] pollutant in the ambient air . . ." 42 U.S.C. § 7408(b). Section 109 (42 U.S.C. 7409) directs the Administrator to propose and promulgate "primary" and "secondary" NAAQS for pollutants for which air quality criteria are issued. Section 109(b)(1) defines a primary standard as one "the attainment and maintenance of which in the judgment of the Administrator, based on such criteria and allowing an adequate margin of safety, are requisite to protect the public health."[1] A

[1] The legislative history of section 109 indicates that a primary standard is to be set at "the maximum permissible ambient air level . . . which will protect the health of any [sensitive] group of the population," and that for this

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secondary standard, as defined in section 109(b)(2), 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." Welfare effects as defined in section 302(h) (42 U.S.C. § 7602(h)) include, but are not limited to, "effects on soils, water, crops, vegetation, man-made materials, animals, wildlife, weather, visibility and climate, damage to and deterioration of property, and hazards to transportation, as well as effects on economic values and on personal comfort and well-being."

In setting standards that are "requisite" to protect public health and welfare, as provided in section 109(b), EPA's task is to establish standards that are neither more nor less stringent than necessary for these purposes. In so doing, EPA may not consider the costs of implementing the standards. See generally, *Whitman v. American Trucking Associations*, 531 U.S. 457, 465-472, 475-76 (2001). Likewise, "[a]ttainability and technological feasibility are not relevant considerations in the promulgation of national ambient air quality standards." *American Petroleum Institute v. Costle*, 665 F. 2d at 1185. Section 109(d)(1) requires that "not later than December 31, 1980, and at 5-year intervals thereafter, the Administrator shall complete a thorough review of the criteria published under section 108 and the national ambient air quality standards . . . and shall make such revisions in such criteria and standards and promulgate such new standards as may be appropriate . . . ." Section 109(d)(2) requires that an independent scientific review committee "shall complete a review of the criteria . . . and the national primary and secondary ambient air quality standards . . . and shall recommend to the Administrator any new . . . standards and revisions of existing criteria and standards as may be appropriate . . . ."

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purpose "reference should be made to a representative sample of persons comprising the sensitive group rather than to a single person in such a group" S. Rep. No. 91-1196, 91st Cong., 2d Sess. 10 (1970).

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Since the early 1980's, this independent review function has been performed by the Clean Air Scientific Advisory Committee (CASAC).

B. History of Reviews of NAAQS for Nitrogen Oxides and Sulfur Oxides

1. NAAQS for Oxides of Nitrogen

After reviewing the relevant science on the public health and welfare effects associated with oxides of nitrogen, EPA promulgated identical primary and secondary NAAQS for NO2 in April 1971. These standards were set at a level of 0.053 parts per million (ppm) as an annual average (36 FR 8186). In 1982, EPA published Air Quality Criteria Document for Oxides of Nitrogen (US EPA, 1982), which updated the scientific criteria upon which the initial standards were based. In February 1984 EPA proposed to retain these standards (49 FR 6866). After taking into account public comments, EPA published the final decision to retain these standards in June 1985 (50 FR 25532).

The EPA began the most recent previous review of the oxides of nitrogen secondary standards in 1987. In November 1991, EPA released an updated draft air quality criteria document (AQCD) for CASAC and public review and comment (56 FR 59285), which provided a comprehensive assessment of the available scientific and technical information on health and welfare effects associated with NO2 and other oxides of nitrogen. The CASAC reviewed the draft document at a meeting held on July 1, 1993 and concluded in a closure letter to the Administrator that the document "provides a scientifically balanced and defensible summary of current knowledge of the effects of this pollutant and provides an adequate basis for EPA to make a decision as to the appropriate NAAQS for NO_2" (Wolff, 1993). The AQCD for Oxides of Nitrogen was then finalized (US EPA, 1995a). The EPA's OAQPS also prepared a Staff Paper that summarized and integrated the key studies and scientific evidence contained in the

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revised AQCD for oxides of nitrogen and identified the critical elements to be considered in the review of the NO_2 NAAQS. The CASAC reviewed two drafts of the Staff Paper and concluded in a closure letter to the Administrator that the document provided a "scientifically adequate basis for regulatory decisions on nitrogen dioxide" (Wolff, 1995).

In October 1995, the Administrator announced her proposed decision not to revise either the primary or secondary NAAQS for NO_2 (60 FR 52874; October 11, 1995). A year later, the Administrator made a final determination not to revise the NAAQS for NO2 after careful evaluation of the comments received on the proposal (61 FR 52852; October 8, 1996). While the primary NO_2 standard was revised in January 2010 by supplementing the existing annual standard with the establishment of a new 1-hour standard, set at a level of 100 ppb (75 FR 6474), the secondary NAAQS for NO_2 remains 0.053 ppm (100 micrograms per cubic meter [µg/m3] of air), annual arithmetic average, calculated as the arithmetic mean of the 1-hour NO2 concentrations.

2. The NAAQS for Oxides of Sulfur

The EPA promulgated primary and secondary NAAQS for SO2 in April 1971 (36 FR 8186). The secondary standards included a standard set at 0.02 ppm, annual arithmetic mean, and a 3-hour average standard set at 0.5 ppm, not to be exceeded more than once per year. These secondary standards were established solely on the basis of evidence of adverse effects on vegetation. In 1973, revisions made to Chapter 5 ("Effects of Sulfur Oxide in the Atmosphere on Vegetation") of the AQCD for Sulfur Oxides (US EPA, 1973) indicated that it could not properly be concluded that the vegetation injury reported resulted from the average SO_2 exposure over the growing season, rather than from short-term peak concentrations. Therefore, EPA proposed (38 FR 11355) and then finalized (38 FR 25678) a revocation of the annual mean secondary

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standard. At that time, EPA was aware that then-current concentrations of oxides of sulfur in the ambient air had other public welfare effects, including effects on materials, visibility, soils, and water. However, the available data were considered insufficient to establish a quantitative relationship between specific ambient concentrations of oxides of sulfur and such public welfare effects (38 FR 25679).

In 1979, EPA announced that it was revising the AQCD for oxides of sulfur concurrently with that for particulate matter (PM) and would produce a combined PM and oxides of sulfur criteria document. Following its review of a draft revised criteria document in August 1980, CASAC concluded that acid deposition was a topic of extreme scientific complexity because of the difficulty in establishing firm quantitative relationships among (1) emissions of relevant pollutants (e.g., SO2 and oxides of nitrogen), (2) formation of acidic wet and dry deposition products, and (3) effects on terrestrial and aquatic ecosystems. The CASAC also noted that acid deposition involves, at a minimum, several different criteria pollutants: oxides of sulfur, oxides of nitrogen, and the fine particulate fraction of suspended particles. The CASAC felt that any document on this subject should address both wet and dry deposition, since dry deposition was believed to account for a substantial portion of the total acid deposition problem.

For these reasons, CASAC recommended that a separate, comprehensive document on acid deposition be prepared prior to any consideration of using the NAAQS as a regulatory mechanism for the control of acid deposition. The CASAC also suggested that a discussion of acid deposition be included in the AQCDs for oxides of nitrogen and PM and oxides of sulfur. Following CASAC closure on the AQCD for oxides of sulfur in December 1981, EPA's OAQPS published a Staff Paper in November 1982, although the paper did not directly assess the issue of acid deposition. Instead, EPA subsequently prepared the following documents to address acid

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deposition: The Acidic Deposition Phenomenon and Its Effects: Critical Assessment Review Papers, Volumes I and II (US EPA, 1984a, b) and The Acidic Deposition Phenomenon and Its Effects: Critical Assessment Document (US EPA, 1985) (53 FR 14935 -14936). These documents, though they were not considered criteria documents and did not undergo CASAC review, represented the most comprehensive summary of scientific information relevant to acid deposition completed by EPA at that point.

In April 1988 (53 FR 14926), EPA proposed not to revise the existing primary and secondary standards for SO_2. This proposed decision with regard to the secondary SO_2 NAAQS was due to the Administrator's conclusions that: (1) based upon the then-current scientific understanding of the acid deposition problem, it would be premature and unwise to prescribe any regulatory control program at that time; and (2) when the fundamental scientific uncertainties had been decreased through ongoing research efforts, EPA would draft and support an appropriate set of control measures. Although EPA revised the primary SO_2 standard in June 2010 by establishing a new 1-hour standard at a level of 75 ppb and revoking the existing 24-hour and annual standards (75 FR 35520), no further decisions on the secondary SO_2 standard have been published.

C. History of Related Assessments and Agency Actions

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

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In spite of the complexities and significant remaining uncertainties associated with the acid deposition problem, it soon became clear that a program to address acid deposition was needed. The Clean Air Act Amendments of 1990 included numerous separate provisions related to the acid deposition problem. The primary and most important of the provisions, the amendments to Title IV of the Act, established the Acid Rain Program to reduce emissions of SO_2 by 10 million tons and emissions of nitrogen oxides by 2 million tons from 1980 emission levels in order to achieve reductions over broad geographic regions. In this provision, Congress included a statement of findings that led them to take action, concluding that (1) the presence of acid compounds and their precursors in the atmosphere and in deposition from the atmosphere represents a threat to natural resources, ecosystems, materials, visibility, and public health; (2) the problem of acid deposition is of national and international significance; and (3) current and future generations of Americans will be adversely affected by delaying measures to remedy the problem.

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

Third, Congress considered that further action might be necessary in the long term to address any problems remaining after implementation of the Title IV program and, reserving judgment on the form that action could take, included Section 404 of the 1990 Amendments (Clean Air Act Amendments of 1990, Pub. L. 101-549, § 404) requiring EPA to conduct a study on the feasibility and effectiveness of an acid deposition standard or standards to protect "sensitive and critically sensitive aquatic and terrestrial resources." At the conclusion of the study, EPA was to submit a report to Congress. Five years later, EPA submitted its report,

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entitled Acid Deposition Standard Feasibility Study: Report to Congress (US EPA, 1995b) in fulfillment of this requirement. That report concluded that establishing acid deposition standards for sulfur and nitrogen deposition may at some point in the future be technically feasible, although appropriate deposition loads for these acidifying chemicals could not be defined with reasonable certainty at that time.

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

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

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criteria pollutants, including impaired visibility, eutrophication of coastal estuaries, global warming, and tropospheric ozone and stratospheric ozone depletion.

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

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

The 2005 NAPAP report states that "… scientific studies indicate that the emission reductions achieved by Title IV are not sufficient to allow recovery of acid-sensitive ecosystems. Estimates from the literature of the scope of additional emission reductions that are necessary in order to protect acid-sensitive ecosystems range from approximately 40-80% beyond full implementation of Title IV...." The results of the modeling presented in this Report to Congress indicate that broader recovery is not predicted without additional emission reductions (NAPAP, 2005).

Given the state of the science as described in the ISA, REA, and in other recent reports, such as the NAPAP reports noted above, EPA has decided, in the context of evaluating the adequacy of the current NO2 and SO2 secondary standards in this review, to revisit the question

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of the appropriateness of setting secondary NAAQS to address remaining known or anticipated adverse public welfare effects resulting from the acidic and nutrient deposition of these criteria pollutants.

D. History of the Current Review

The EPA initiated this current review in December 2005 with a call for information (70 FR 73236) for the development of a revised ISA. An Integrated Review Plan (IRP) was developed to provide the framework and schedule as well as the scope of the review and to identify policy-relevant questions to be addressed in the components of the review. The IRP was released in 2007 (US EPA, 2007) for CASAC and public review. The EPA held a workshop in July 2007 on the ISA to obtain broad input from the relevant scientific communities. This workshop helped to inform the preparation of the first draft ISA, which was released for CASAC and public review in December 2007; a CASAC meeting was held on April 2-3, 2008 to review the first draft ISA. A second draft ISA was released for CASAC and public review in August 2008, and was discussed at a CASAC meeting held on October 1-2, 2008. The final ISA (US EPA, 2008) was released in December 2008.

Based on the science presented in the ISA, EPA developed the REA to further assess the national impact of the effects documented in the ISA. The Draft Scope and Methods Plan for Risk/Exposure Assessment: Secondary NAAQS Review for Oxides of Nitrogen and Oxides of Sulfur outlining the scope and design of the future REA was prepared for CASAC consultation and public review in March 2008. A first draft REA was presented to CASAC and the public for review in August 2008 and a second draft was presented for review in June 2009. The final REA (US EPA, 2009) was released in September 2009. A first draft PA was released in March 2010 and reviewed by CASAC on April 1-2, 2010. In a June 22, 2010 letter to the Administrator,

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CASAC provided advice and recommendations to the Agency concerning the first draft PA (Russell and Samet, 2010a). A second draft PA was released to CASAC and the public in September 2010 and reviewed by CASAC on October 6-7, 2010. The CASAC provided advice and recommendations to the Agency regarding the second draft PA in a December 9, 2010 letter (Russell and Samet 2010b). The CASAC and public comments on the second draft PA were considered by EPA staff in developing a final PA (US EPA, 2011). CASAC requested an additional meeting to provide additional advice to the Administrator based on the final PA on February 15–16, 2011. On January 14, 2011, EPA released a version of the final PA prior to final document production, to provide sufficient time for CASAC review of the document in advance of this meeting. The final PA, incorporating final reference checks and document formatting, was released in February 2011.. In a May 17, 2011 letter (Russell and Samet, 2011a), CASAC offered additional advice and recommendations to the Administrator with regard to the review of the secondary NAAQS for oxides of nitrogen and oxides of sulfur.

In 2005, the Center for Biological Diversity and four other plaintiffs filed a complaint alleging that EPA had failed to complete the current review within the period provided by statute.[2] The schedule for completion of this review is governed by a consent decree resolving that lawsuit and the subsequent extension agreed to by the parties. The schedule presented in the original consent decree that governs this review, entered by the court on November 19, 2007, was revised on October 22, 2009 to allow for a 17-month extension of the schedule. The current decree provides that EPA sign for publication notices of proposed and final rulemaking concerning its review of the oxides of nitrogen and oxides of sulfur NAAQS no later than July 12, 2011 and March 20, 2012, respectively.

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[2] *Center for Biological Diversity, et al. v. Johnson, No. 05–1814 (D.D.C.)*

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This action presents the Administrator's proposed decisions on the review of the current secondary oxides of nitrogen and oxides of sulfur standards. Throughout this preamble a number of conclusions, findings, and determinations proposed by the Administrator are noted. While they identify the reasoning that supports this proposal, they are only proposals and are not intended to be final or conclusive in nature. The EPA invites general, specific, and/or technical comments on all issues involved with this proposal, including all such proposed judgments, conclusions, findings, and determinations.

E. Scope of the Current Review

In conducting this periodic review of the secondary NAAQS for oxides of nitrogen and oxides of sulfur, as discussed in the IRP and REA, EPA decided to assess the scientific information, associated risks, and standards relevant to protecting the public welfare from adverse effects associated jointly with oxides of nitrogen and sulfur. Although EPA has historically adopted separate secondary standards for oxides of nitrogen and oxides of sulfur, EPA is conducting a joint review of these standards because oxides of nitrogen and sulfur, and their associated transformation products are linked from an atmospheric chemistry perspective, as well as from an environmental effects perspective. The National Research Council (NRC) has recommended that EPA consider multiple pollutants, as appropriate, in forming the scientific basis for the NAAQS (NRC, 2004). As discussed in the ISA and REA, there is a strong basis for considering these pollutants together, building upon EPA's past recognition of the interactions of these pollutants and on the growing body of scientific information that is now available related to these interactions and associated ecological effects.

In defining the scope of this review, it must be considered that EPA has set secondary standards for two other criteria pollutants related to oxides of nitrogen and sulfur: ozone and

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particulate matter (PM). Oxides of nitrogen are precursors to the formation of ozone in the atmosphere, and under certain conditions, can combine with atmospheric ammonia to form ammonium nitrate, a component of fine PM. Oxides of sulfur are precursors to the formation of particulate sulfate, which is a significant component of fine PM in many parts of the U.S. There are a number of welfare effects directly associated with ozone and fine PM, including ozone-related damage to vegetation and PM-related visibility impairment. Protection against those effects is provided by the ozone and fine PM secondary standards. This review focuses on evaluation of the protection provided by secondary standards for oxides of nitrogen and sulfur for two general types of effects: (1) direct effects on vegetation associated with exposure to gaseous oxides of nitrogen and sulfur in the ambient air, which are the effects that the current NO_2 and SO_2 secondary standards protect against; and (2) effects associated with the deposition of oxides of nitrogen and sulfur to sensitive aquatic and terrestrial ecosystems, including deposition in the form of particulate nitrate and particulate sulfate.

The ISA focuses on the ecological effects associated with deposition of ambient oxides of nitrogen and sulfur to natural sensitive ecosystems, as distinguished from commercially managed forests and agricultural lands. This focus reflects the fact that the majority of the scientific evidence regarding acidification and nutrient enrichment is based on studies in unmanaged ecosystems. Non-managed terrestrial ecosystems tend to have a higher fraction of nitrogen deposition resulting from atmospheric nitrogen (US EPA, 2008, section 3.3.2.5). In addition, the ISA notes that agricultural and commercial forest lands are routinely fertilized with amounts of nitrogen that exceed air pollutant inputs even in the most polluted areas (US EPA, 2008, section 3.3.9). This review recognizes that the effects of nitrogen deposition in managed areas are viewed differently from a public welfare perspective than are the effects of nitrogen deposition in

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natural, unmanaged ecosystems, largely due to the more homogeneous, controlled nature of species composition and development in managed ecosystems and the potential for benefits of increased productivity in those ecosystems.

In focusing on natural sensitive ecosystems, the PA primarily considers the effects of ambient oxides of nitrogen and sulfur via deposition on multiple ecological receptors. The ISA highlights effects including those associated with acidification and nitrogen nutrient enrichment. With a focus on these deposition-related effects, EPA's objective is to develop a framework for oxides of nitrogen and sulfur standards that incorporates ecologically relevant factors and that recognizes the interactions between the two pollutants as they deposit to sensitive ecosystems. The overarching policy objective is to develop a secondary standard(s) based on the ecological criteria described in the ISA and the results of the assessments in the REA, and consistent with the requirement of the CAA to set secondary standards that are requisite to protect the public welfare from any known or anticipated adverse effects associated with the presence of these air pollutants in the ambient air. Consistent with the CAA, this policy objective includes consideration of "variable factors . . . which of themselves or in combination with other factors may alter the effects on public welfare" of the criteria air pollutants included in this review.

In addition, we have chosen to focus on the effects of ambient oxides of nitrogen and sulfur on ecological impacts on sensitive aquatic ecosystems associated with acidifying deposition of nitrogen and sulfur, which is a transformation product of ambient oxides of nitrogen and sulfur. Based on the information in the ISA, the assessments presented in the REA, and advice from CASAC on earlier drafts of this PA (Russell and Samet, 2010a, 2010b), and as discussed in detail in the PA, we have the greatest confidence in the causal linkages between

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oxides of nitrogen and sulfur and aquatic acidification effects relative to other deposition-related effects, including terrestrial acidification and aquatic and terrestrial nutrient enrichment.

II. Rationale for Proposed Decision on the Adequacy of the Current Secondary Standards

Decisions on retaining or revising the current secondary standards for oxides of nitrogen and sulfur are largely public welfare policy judgments based on the Administrator's informed assessment of what constitutes requisite protection against adverse effects to public welfare. A public welfare policy decision should draw upon scientific information and analyses about welfare effects, exposure and risks, as well as judgments about the appropriate response to the range of uncertainties that are inherent in the scientific evidence and analyses. The ultimate determination as to what level of damage to ecosystems and the services provided by those ecosystems is adverse to public welfare is not wholly a scientific question, although it is informed by scientific studies linking ecosystem damage to losses in ecosystem services, and information on the value of those losses of ecosystem services. In reaching such decisions, the Administrator seeks to establish standards that are neither more nor less stringent than necessary for this purpose.

This section presents the rationale for the Administrator's proposed conclusions with regard to the adequacy of protection and ecological relevance of the current secondary standards for oxides of nitrogen and sulfur. As discussed more fully below, this rationale considered the latest scientific information on ecological effects associated with the presence of oxides of nitrogen and oxides of sulfur in the ambient air. This rationale also takes into account: (1) staff assessments of the most policy-relevant information in the ISA and staff analyses of air quality, exposure, and ecological risks, presented more fully in the REA and in the PA, upon which staff

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conclusions on revisions to the secondary oxides of nitrogen and oxides of sulfur standards are based; (2) CASAC advice and recommendations, as reflected in discussions of drafts of the ISA, REA, and PA at public meetings, in separate written comments, and in CASAC's letters to the Administrator; and (3) public comments received during the development of these documents, either in connection with CASAC meetings or separately.

In developing this rationale, EPA has drawn upon an integrative synthesis of the entire body of evidence, published through early 2008, on ecological effects associated with the deposition of oxides of nitrogen and oxides of sulfur in the ambient air (US EPA, 2008). As discussed below in section II.A, this body of evidence addresses a broad range of ecological endpoints associated with ambient levels of oxides of nitrogen and oxides of sulfur. In considering this evidence, EPA focuses on those ecological endpoints, such as aquatic acidification, for which the ISA judges associations with oxides of nitrogen and oxides of sulfur to be causal, likely causal, or for which the evidence is suggestive that oxides of nitrogen and/or sulfur contribute to the reported effects. The categories of causality determinations have been developed in the ISA (US EPA, 2008) and are discussed in Section 1.6 of the ISA.

Crucial to this review is the development of a form for an ecologically relevant standard that reflects both the geographically variable and deposition-dependent nature of the effects. The atmospheric levels of oxides of nitrogen and sulfur that afford a particular level of ecosystem protection are those levels that result in an amount of deposition that is less than the amount of deposition that a given ecosystem can accept without defined levels of degradation.

Drawing from the framework developed in the REA, the framework we used to structure an ecologically meaningful secondary standard in the PA and to further develop the indicator, form, level, and averaging time of such a standard in section III of this proposal is depicted

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below and highlights the three key linkages that need to be considered in developing an ecologically relevant standard.

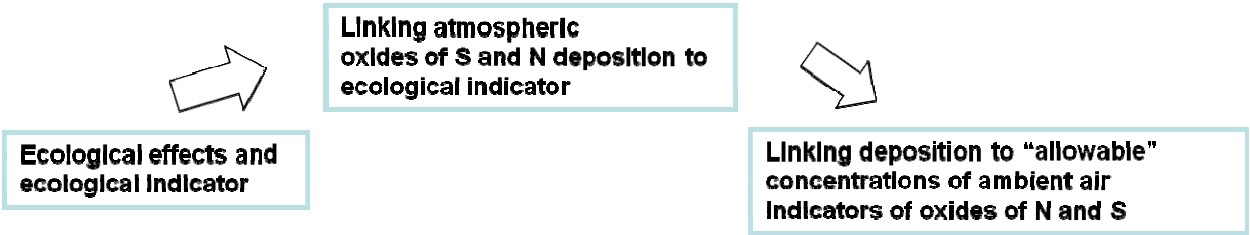

Figure II-1. Simplified conceptual design of the form of an aquatic acidification standard for oxides of nitrogen and sulfur.

The following discussion relies heavily on chapters 2 and 3 of the PA. The PA includes staff's evaluation of the policy implications of the scientific assessment of the evidence presented and assessed in the ISA and the results of quantitative assessments based on that information presented and assessed in the REA. Taken together, this information informs staff conclusions and the development of policy options in the PA for consideration in addressing public and welfare effects associated with the presence of oxides of nitrogen and oxides of sulfur in the ambient air. Of particular note, chapter 2 of the PA presents information not repeated here that characterizes emissions, air quality, deposition and water quality. It includes discussions of the sources of nitrogen and sulfur in the atmosphere as well as current ambient air quality monitoring networks and models. Additional information in this section includes ecological modeling and water quality data sources.

Section II.A presents a discussion of the effects associated with oxides of nitrogen and sulfur in the ambient air. The discussion is organized around the types of effects being

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considered, including direct effects of gaseous oxides of nitrogen and sulfur, deposition-related effects related to acidification and nutrient enrichment, and other effects such as materials damage, climate-related effects and mercury methylation.

Section II.B presents a summary and discussion of the risk and exposure assessment performed for each of the four major effects categories. The REA uses case studies representing the broad geographic variability of the impacts from oxides of nitrogen and sulfur to conclude that there are ongoing adverse effects in many ecosystems from deposition of oxides of nitrogen and sulfur and that under current emissions scenarios these effects are likely to continue.

Section II.C presents a discussion of adversity linking ecological effects to measures that can be used to characterize the extent to which such effects are reasonably considered to be adverse to public welfare. This involves consideration of how to characterize adversity from a public welfare perspective. In so doing, consideration is given to the concept of ecosystem services, the evidence of effects on ecosystem services, and how ecosystem services can be linked to ecological indicators.

Section II.D presents an assessment of the adequacy of the current oxides of nitrogen and oxides of sulfur secondary standards. Consideration is given to the adequacy of protection afforded by the current standards for both direct and deposition-related effects, as well as to the appropriateness of the fundamental structure and the basic elements of the current standards for providing protection from deposition-related effects. Considerations as to the extent to which deposition-related effects that could reasonably be judged to be adverse to public welfare are occurring under current conditions which are allowed by the current standards is also considered. Discussion of the structures and basic elements of the current NO_2 and SO_2 secondary standards and whether they are adequate to protect against such effects is presented.

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A. *Ecological Effects*

This section discusses the known or anticipated ecological effects associated with oxides of nitrogen and sulfur, including the direct effects of gas-phase exposure to oxides of nitrogen and sulfur (section II.A.1) and effects associated with deposition-related exposure (sections II.A.2 and 3). Section II.A. 2 addresses effects related to acidification of aquatic and terrestrial ecosystems and section II A.3 addresses effects related to nutrient enrichment of aquatic and terrestrial ecosystems. These sections also address questions about the nature and magnitude of ecosystem responses to reactive nitrogen and sulfur deposition, including responses related to acidification, nutrient depletion, and, in Section II.A 4 the mobilization of toxic metals in sensitive aquatic and terrestrial ecosystems. The uncertainties and limitations associated with the evidence of such effects are also discussed throughout this section.

1. Effects Associated with Gas-Phase Oxides of Nitrogen and Sulfur

Ecological effects on vegetation as discussed in earlier reviews as well as the ISA can be attributed to gas-phase oxides of nitrogen and sulfur. Acute and chronic exposures to gaseous pollutants such as SO_2, NO_2, nitric oxide (NO), nitric acid (HNO_3) and peroxyacetyl nitrite (PAN) are associated with negative impacts to vegetation. The current secondary NAAQS were set to protect against direct damage to vegetation by exposure to gas-phase oxides of nitrogen and sulfur, such as foliar injury, decreased photosynthesis, and decreased growth. The following summary is a concise overview of the known or anticipated effects to vegetation caused by gas phase nitrogen and sulfur. Most phototoxic effects associated with gas phase oxides of nitrogen and sulfur occur at levels well above ambient concentrations observed in the U.S. (US EPA, 2008, section 3.4.2.4).

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a. Nature of ecosystem responses to gas-phase nitrogen and sulfur

The 2008 ISA found that gas phase nitrogen and sulfur are associated with direct phytotoxic effects (US EPA, 2008, section 4.4). The evidence is sufficient to infer a causal relationship between exposure to SO_2 and injury to vegetation (US EPA, 2008, section 4.4.1 and 3.4.2.1). Acute foliar injury to vegetation from SO_2 may occur at levels above the current secondary standard (3-h average of 0.50 ppm). Effects on growth, reduced photosynthesis and decreased yield of vegetation are also associated with increased SO_2 exposure concentration and time of exposure.

The evidence is sufficient to infer a causal relationship between exposure to NO, NO_2 and PAN and injury to vegetation (US EPA, 2008, section 4.4.2 and 3.4.2.2). At sufficient concentrations, NO, NO_2 and PAN can decrease photosynthesis and induce visible foliar injury to plants. Evidence is also sufficient to infer a causal relationship between exposure to HNO_3 and changes to vegetation (US EPA, 2008, section 4.4.3 and 3.4.2.3). Phytotoxic effects of this pollutant include damage to the leaf cuticle in vascular plants and disappearance of some sensitive lichen species.

b. Magnitude of ecosystem response to gas-phase nitrogen and sulfur

Vegetation in ecosystems near sources of gaseous oxides of nitrogen and sulfur or where SO_2, NO, NO_2, PAN and HNO_3 are most concentrated are more likely to be impacted by these pollutants. Uptake of these pollutants in a plant canopy is a complex process involving adsorption to surfaces (leaves, stems and soil) and absorption into leaves (US EPA, 2008, section 3.4.2). The functional relationship between ambient concentrations of gas phase oxides of nitrogen and sulfur and specific plant response are impacted by internal factors such as rate of stomatal conductance and plant detoxification mechanisms, and external factors including plant

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water status, light, temperature, humidity, and pollutant exposure regime (US EPA, 2008, section 3.4.2).

Entry of gases into a leaf is dependent upon physical and chemical processes of gas phase as well as to stomatal aperture. The aperture of the stomata is controlled largely by the prevailing environmental conditions, such as water availability, humidity, temperature, and light intensity. When the stomata are closed, resistance to gas uptake is high and the plant has a very low degree of susceptibility to injury. Mosses and lichens do not have a protective cuticle barrier to gaseous pollutants or stomata and are generally more sensitive to gaseous sulfur and nitrogen than vascular plants (US EPA, 2008, section 3.4.2).

The appearance of foliar injury can vary significantly across species and growth conditions affecting stomatal conductance in vascular plants (US EPA, 2009, section 6.4.1). For example, damage to lichens from SO_2 exposure includes decreased photosynthesis and respiration, damage to the algal component of the lichen, leakage of electrolytes, inhibition of nitrogen fixation, decreased potassium (K+) absorption, and structural changes.

The phytotoxic effects of gas phase oxides of nitrogen and sulfur are dependent on the exposure concentration and duration and species sensitivity to these pollutants. Effects to vegetation associated with oxides of nitrogen and sulfur are therefore variable across the U.S. and tend to be higher near sources of photochemical smog. For example, SO_2 is considered to be the primary factor contributing to the death of lichens in many urban and industrial areas.

The ISA states there is very limited new research on phytotoxic effects of NO, NO_2, PAN and HNO_3 at concentrations currently observed in the U.S. with the exception of some lichen species (US EPA, 2008, section 4.4). Past and current HNO_3 concentrations may be contributing to the decline in lichen species in the Los Angeles basin. Most phytotoxic effects associated

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with gas phase oxides of nitrogen and sulfur occur at levels well above ambient concentrations observed in the U.S. (US EPA, 2008, section 3.4.2.4).

2. Acidification Effects Associated with Deposition of Oxides of Nitrogen and Sulfur

Sulfur oxides and nitrogen oxides in the atmosphere undergo a complex mix of reactions in gaseous, liquid, and solid phases to form various acidic compounds. These acidic compounds are removed from the atmosphere through deposition: either wet (e.g., rain, snow), fog or cloud, or dry (e.g., gases, particles). Deposition of these acidic compounds to ecosystems can lead to effects on ecosystem structure and function. Following deposition, these compounds can, in some instances, unless retained by soil or biota, leach out of the soils in the form of sulfate (SO_4^{2-}) and nitrate (NO_3^-), leading to the acidification of surface waters. The effects on ecosystems depend on the magnitude and rate of deposition, as well as a host of biogeochemical processes occurring in the soils and water bodies (US EPA, 2009, section 2.1). The chemical forms of nitrogen that may contribute to acidifying deposition include both oxidized and reduced chemical species, including reduced forms of nitrogen (NH_x).

When sulfur or nitrogen leaches from soils to surface waters in the form of SO_4^{2-} or NO_3^-, an equivalent amount of positive cations, or countercharge, is also transported. This maintains electroneutrality. If the countercharge is provided by base cations, such as calcium (Ca^{2+}), magnesium (Mg^{2+}), sodium (Na^+), or K^+, rather than hydrogen (H^+) and dissolved inorganic aluminum, the acidity of the soil water is neutralized, but the base saturation of the soil decreases. Continued SO_4^{2-} or NO_3^- leaching can deplete the available base cation pool in soil. As the base cations are removed, continued deposition and leaching of SO_4^{2-} and/or NO_3^- (with H^+ and Al^{3+}) leads to acidification of soil water, and by connection, surface water. Introduction of strong acid anions such as sulfate and nitrate to an already acidic soil, whether naturally or due

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to anthropogenic activities, can lead to instantaneous acidification of waterbodies through direct runoff without any significant change in base cation saturation. The ability of a watershed to neutralize acidic deposition is determined by a variety of biogeophysical factors including weathering rates, bedrock composition, vegetation and microbial processes, physical and chemical characteristics of soils and hydrologic flowpaths (US EPA, 2009, section 2.1). Some of these factors such as vegetation and soil depth are highly variable over small spatial scales such as meters, but can be aggregated to evaluate patterns over larger spatial scales. Acidifying deposition of oxides of nitrogen and sulfur and the chemical and biological responses associated with these inputs vary temporally. Chronic or long-term deposition processes in the time scale of years to decades result in increases in inputs of nitrogen and sulfur to ecosystems and the associated ecological effects. Episodic or short term (i.e., hours or days) deposition refers to events in which the level of the acid neutralizing capacity (ANC) of a lake or stream is temporarily lowered. In aquatic ecosystems, short-term (i.e., hours or days) episodic changes in water chemistry can have significant biological effects. Episodic acidification refers to conditions during precipitation or snowmelt events when proportionately more drainage water is routed through upper soil horizons that tend to provide less acid neutralizing than is passing through deeper soil horizons (US EPA, 2009, section 4.2). In addition, the accumulated sulfate and nitrate in snow packs can provide a surge of acidic inputs. Some streams and lakes may have chronic or base flow chemistry that is suitable for aquatic biota, but may be subject to occasional acidic episodes with deleterious consequences to sensitive biota.

The following summary is a concise overview of the known or anticipated effects caused by acidification to ecosystems within the U.S. Acidification affects both terrestrial and freshwater aquatic ecosystems.

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a. Nature of acidification-related ecosystem responses

The ISA concluded that deposition of oxides of nitrogen and sulfur and NH_x leads to the varying degrees of acidification of ecosystems (US EPA, 2008). In the process of acidification, biogeochemical components of terrestrial and freshwater aquatic ecosystems are altered in a way that leads to effects on biological organisms. Deposition to terrestrial ecosystems often moves through the soil and eventually leaches into adjacent water bodies.

i. Aquatic ecosystems

The scientific evidence is sufficient to infer a causal relationship between acidifying deposition and effects on biogeochemistry and biota in aquatic ecosystems (US EPA, 2008, section 4.2.2). The strongest evidence comes from studies of surface water chemistry in which acidic deposition is observed to alter sulfate and nitrate concentrations in surface waters, the sum of base cations, ANC, dissolved inorganic aluminum and pH (US EPA, 2008, section 3.2.3.2). The ANC is a key indicator of acidification with relevance to both terrestrial and aquatic ecosystems. The ANC is useful because it integrates the overall acid-base status of a lake or stream and reflects how aquatic ecosystems respond to acidic deposition over time. There is also a relationship between ANC and the surface water constituents that directly contribute to or ameliorate acidity-related stress, in particular, concentrations of hydrogen ion (as pH), Ca^{2+} and aluminum (Al). Moreover, low pH surface waters leach aluminum from soils, which is quite lethal to fish and other aquatic organisms. In aquatic systems, there is a direct relationship between ANC and fish and phyto-zooplankton diversity and abundance.

Low ANC coincides with effects on aquatic systems (e.g., individual species fitness loss or death, reduced species richness, altered community structure). At the community level, species richness is positively correlated with pH and ANC because energy cost in maintaining

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physiological homeostasis, growth, and reproduction is high at low ANC levels. For example, there is a logistic relationship between fish species richness and ANC class for Adirondack Case Study Area lakes that indicates the probability of occurrence of an organism for a given value of ANC. Biota are generally not harmed when ANC values are >100 microequivalents per liter (μeq/L). The number of fish species also peaks at ANC values >100 μeq/L. Below 100 μeq/L ANC, fish fitness and community diversity begin to decline (US EPA, section 4.2). Specifically at ANC levels between 100 and 50 μeq/L, the fitness of sensitive species (e.g., brook trout, zooplankton) begins to decline. When ANC concentrations are <50 μeq/L, they are generally associated with death or loss of fitness of biota that are sensitive to acidification.

Consistent and coherent documentation from multiple studies on various species from all major trophic levels of aquatic systems shows that geochemical alteration caused by acidification can result in the loss of acid-sensitive biological species (US EPA, 2008, section 3.2.3.3). This is most often discussed with relation to pH. For example, in the Adirondacks, of the 53 fish species recorded in Adirondack lakes about half (26 species) were absent from lakes with pH below 6.0. Biological effects are linked to changes in water chemistry including decreases in ANC and pH and increases in inorganic Al concentration. The direct biological effects are caused by lowered pH which leads to increased inorganic Al concentrations (US EPA, 2011, Figures 3-1 and 3-2). While ANC level does not cause direct biological harm it is a good overall indicator of the risk of acidification (US EPA, 2011, section 3.1.3).

There are clear associations between ANC, pH and aquatic species mortality and health which are summarized in section 3.1.1 of the PA. Significant harm to sensitive aquatic species has been observed at pH levels below 6. Normal stream pH levels with little to no toxicity range from 6 to 7 (MacAvoy et al, 1995). Baker et al (1990) observed that "lakes with pH less than

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approximately 6.0 contain significantly fewer species than lakes with pH levels above 6.0." As noted in Chapter 3, typically at pH <4.5 and an ANC <0 μeq/L, complete to near-complete loss of many taxa of organisms occur, including fish and aquatic insect populations, whereas other taxa are reduced to only acidophilic species. Acid Neutralizing Capacity is a measure of how much acid can be neutralized in a specific surface water system. An ANC value of 0 or below means that surface waters have no ability to neutralize any additional acid inputs.

Additional evidence can help refine the understanding of effects occurring at pH levels between 4.5 and 6. When pH levels are below 5.6, relatively lower trout survival rates were observed in the Shenandoah National Park. In field observations, when pH levels dropped to 5, mortality rates went to 100 percent (Bulger et al, 2000). At pH levels ranging from 5.4 to 5.8, cumulative mortality continues to increase. Several studies have shown that trout exposed to water with varying pH levels and fish larvae showed increasing mortality as pH levels decrease. In one study almost 100 percent mortality was observed at a pH of 4.5 compared to almost 100 percent survival at a pH of 6.5. Intermediate pH values (6.0, 5.5) in all cases showed reduced survival compared with the control (6.5), but not by statistically significant amounts (US EPA, 2008, section 3.2.3.3).

One important indicator of acid stress is increased fish mortality. The response of fish to pH is not uniform across species. A number of synoptic surveys indicated loss of species diversity and absence of several fish species in the pH range of 5.0 to 5.5. If pH is lower, there is a greater likelihood that more fish species could be lost without replacement, resulting in decreased richness and diversity. In general, populations of salmonids are not found at pH levels less than 5.0, and smallmouth bass (*Micropterus dolomieu*) populations are usually not found at pH values less than about 5.2 to 5.5. From Table 3-1, only one study showed significant

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mortality effects above a pH of 6, while a number of studies showed significant mortality when pH levels are at or below 5.5.

The highest pH level for any of the studies reported in the ISA is 6.0, suggesting that pH above 6.0 is protective against mortality effects for most species. Most thresholds are in the range of pH of 5.0 to 6.0, which suggests that a target pH should be no lower than 5.0. Protection against mortality in some recreationally important species such as lake trout (pH threshold of 5.6) and crappie (pH threshold of 5.5), combined with the evidence of effects on larval and embryo survival suggests that pH levels greater than 5.5 should be targeted to provide protection against mortality effects throughout the life stages of fish.

Non-lethal effects have been observed at pH levels as high as 6. A study in the Shenandoah National Park found that the condition factor, a measure of fish health expressed as fish weight/length multiplied by a scaling constant, is positively correlated with stream pH levels, and that the condition factor is reduced in streams with a pH of 6.0 (US EPA, 2008, section 3.2.3.3).

Biodiversity is another indicator of aquatic ecosystem health. A key study in the Adirondacks found that lakes with a pH of 6.0 had only half the potential species of fish (27 of 53 potential species). There is often a positive relationship between pH and number of fish species, at least for pH values between about 5.0 and 6.5, or ANC values between about 0 to 100 μeq/L. Such observed relationships are complicated, however, by the tendency for smaller lakes and streams, having smaller watersheds, to also support fewer fish species, irrespective of acid-base chemistry. This pattern may be due to a decrease in the number of available niches as stream or lake size decreases. Nevertheless, fish species richness is relatively easily determined and is one of the most useful indicators of biological effects of surface water acidification.

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Changes in stream water pH and ANC also contribute to declines in taxonomic richness of zooplankton, and macroinvertebrates which are often sources of food for fish, birds and other animal species in various ecosystems. These fish may also serve as a source of food and recreation for humans. Acidification of ecosystems has been shown to disrupt food web dynamics causing alteration to the diet, breeding distribution, and reproduction of certain species of birds (US EPA, 2008, section 4.2.2.2. and Table 3-9). For example, breeding distribution of the common goldeneye (*Bucephala clangula*), an insectivorous duck, may be affected by changes in acidifying deposition. Similarly, decreases in prey diversity and quantity have been observed to create feeding problems for nesting pairs of loons on low-pH lakes in the Adirondacks.

ii. Terrestrial ecosystems

In terrestrial ecosystems, the evidence is sufficient to infer a causal relationship between acidifying deposition and changes in biogeochemistry (US EPA, 2008, section 4.2.1.1). The strongest evidence comes from studies of forested ecosystems, with supportive information on other plant taxa, including shrubs and lichens (US EPA, 2008, section 3.2.2.1.). Three useful indicators of chemical changes and acidification effects on terrestrial ecosystems, showing consistency and coherence among multiple studies are: soil base saturation, Al concentrations in soil water, and soil carbon to nitrogen (C:N) ratio (US EPA, 2008, section 3.2.2.2).

As discussed in the ISA and REA, in soils with base saturation less than about 15 to 20 percent, exchange chemistry is dominated by Al. Under these conditions, responses to inputs of sulfuric acid and HNO_3 largely involve the release and mobilization of dissolved inorganic Al. The effect can be neutralized by weathering from geologic parent material or base cation exchange. The Ca^{2+} and Al concentrations in soil water are strongly influenced by soil

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acidification and both have been shown to have quantitative links to tree health, including Al interference with Ca^{2+} uptake and Al toxicity to roots. Effects of nitrification and associated acidification and cation leaching have been consistently shown to occur only in soils with a C:N ratio below about 20 to 25.

Soil acidification caused by acidic deposition has been shown to cause decreased growth and increased susceptibility to disease and injury in sensitive tree species. Red spruce (*Picea rubens*) dieback or decline has been observed across high elevation areas in the Adirondack, Green and White mountains. The frequency of freezing injury to red spruce needles has increased over the past 40 years, a period that coincided with increased emissions of sulfur and nitrogen oxides and increased acidifying deposition. Acidifying deposition can contribute to dieback in sugar maple (*Acer saccharum*) through depletion of cations from soil with low levels of available Ca. Grasslands are likely less sensitive to acidification than forests due to grassland soils being generally rich in base cations.

iii. Ecosystem sensitivity

The intersection between current deposition loading, historic loading and sensitivity defines the ecological vulnerability to the effects of acidification. Freshwater aquatic and some terrestrial ecosystems, notably forests, are the ecosystem types which are most sensitive to acidification. The ISA reports that the principal factor governing the sensitivity of terrestrial and aquatic ecosystems to acidification from sulfur and nitrogen deposition is geology (particularly surficial geology). Geologic formations having low base cation supply generally underlie the watersheds of acid-sensitive lakes and streams. Other factors that contribute to the sensitivity of soils and surface waters to acidifying deposition include topography, soil chemistry, land use,

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and hydrologic flowpaths. Episodic and chronic acidification tends to occur in areas that have base-poor bedrock, high relief, and shallow soils (US EPA, 2008, section 3.2.4.1).

b. Magnitude of acidification-related ecosystem responses

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

i. Aquatic acidification

Freshwater ecosystem surveys and monitoring in the eastern U.S. have been conducted by many programs since the mid-1980s, including EPA's Environmental Monitoring and Assessment Program (EMAP), National Surface Water Survey (NSWS), Temporally Integrated Monitoring of Ecosystems (TIME), and Long-term Monitoring (LTM) programs. Based on analyses of surface water data from these programs, New England, the Adirondack Mountains, the Appalachian Mountains (northern Appalachian Plateau and Ridge/Blue Ridge region) and the Upper Midwest contain the most sensitive lakes and streams (i.e., ANC less than about 50 μeq/L). Portions of northern Florida also contain many acidic and low-ANC lakes and streams, although the role of acidifying deposition in this region is less clear. The western U.S. contains many of the surface waters most sensitive to potential acidification effects, but with the exception of the Los Angeles Basin and surrounding areas, the levels of acidifying deposition are low in most areas. Therefore, acidification of surface waters by acidic deposition is not as prevalent in the western U.S., and the extent of chronic surface water acidification that has occurred in that region to date has likely been very limited relative to the Eastern U.S. (US EPA, 2008, section 3.2.4.2 and US EPA, 2009, section 4.2.2).

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There are a number of species including fish, aquatic insects, other invertebrates and algae that are sensitive to acidification and cannot survive, compete or reproduce in acidic waters (US EPA, 2008, section 3.2.3.3). Decreases in ANC and pH have been shown to contribute to declines in species richness and declines in abundance of zooplankton, macroinvertebrates, and fish. Reduced growth rates have been attributed to acid stress in a number of fish species including Atlantic salmon (*Salmo salar*), Chinook salmon (*Oncorhynchus tshawytscha*), lake trout (*Salvelinus namaycush*), rainbow trout (*Oncorhynchis mykiss*), brook trout (*Salvelinus Fontinalis*), and brown trout (*Salmo trutta*). In response to small to moderate changes in acidity, acid-sensitive species are often replaced by other more acid-tolerant species, resulting in changes in community composition and richness. The effects of acidification are continuous, with more species being affected at higher degrees of acidification. At a point, typically a pH <4.5 and an ANC <0 μeq/L, complete to near-complete loss of many taxa of organisms occur, including fish and aquatic insect populations, whereas other taxa are reduced to only acidophilic species. These changes in taxa composition are associated with the high energy cost in maintaining physiological homeostasis, growth, and reproduction at low ANC levels (US EPA, 2008, section 3.2.3.3). Decreases in species richness related to acidification have been observed in the Adirondack Mountains and Catskill Mountains of New York, New England and Pennsylvania, and Virginia. From the sensitive areas identified by the ISA, further "case study" analyses on aquatic ecosystems in the Adirondack Mountains and Shenandoah National Park were conducted to better characterize ecological risk associated with acidification (US EPA, 2009, section 4).

The ANC is the most widely used indicator of acid sensitivity and has been found in various studies to be the best single indicator of the biological response and health of aquatic communities in acid-sensitive systems (Lien et al., 1992; Sullivan et al., 2006; US EPA, 2008).

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In the REA, surface water trends in SO_4^{2-} and NO_3^- concentrations and ANC levels were analyzed to affirm the understanding that reductions in deposition could influence the risk of acidification. The ANC values have been categorized according to their effects on biota, as shown in the table below. Monitoring data from TIME/LTM and EMAP programs were assessed for the years 1990 to 2006, and past, present and future water quality levels were estimated by both steady-state and dynamic biogeochemical models.

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Table II-1. Ecological effects associated with alternative levels of acid neutralizing capacity (ANC). (source: USEPA, Acid Rain Program)

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

Studies on fish species richness in the Adirondacks Case Study Area demonstrated the effect of acidification. Of the 53 fish species recorded in Adirondack Case Study Area lakes, only 27 species were found in lakes with a pH <6.0. The 26 species missing from lakes with a pH <6.0 include important recreational species, such as Atlantic salmon, tiger trout (*Salmo trutta* X *Salvelinus fontinalis*), redbreast sunfish (*Lepomis auritus*), bluegill (*Lepomis macrochirus*),

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tiger musky (*Esox masquinongy* X *lucius*), walleye (*Sander vitreus*), alewife (*Alosa pseudoharengus*), and kokanee (*Oncorhynchus nerka*), as well as ecologically important minnows that are commonly consumed by sport fish. A survey of 1,469 lakes in the late 1980s found 346 lakes to be devoid of fish. Among lakes with fish, there was a relationship between the number of fish species and lake pH, ranging from about one species per lake for lakes having a pH <4.5 to about six species per lake for lakes having a pH >6.5. In the Adirondacks, a positive relationship exists between the pH and ANC in lakes and the number of fish species present in those lakes (US EPA, 2008, section 3.2.3.4).

Since the mid-1990s, streams in the Shenandoah Case Study Area have shown slight declines in NO_3^- and SO_4^{2-} concentrations in surface waters. The 2006 concentrations are still above pre-acidification (1860) conditions. Model of Acidification of Groundwater in Catchments (MAGIC) modeling predicts surface water concentrations of NO_3^- and SO_4^{2-} are10- and 32-fold higher, respectively, in 2006 than in 1860. The estimated average ANC across 60 streams in the Shenandoah Case Study Area is 57.9 μeq/L (± 4.5 μeq/L). Fifty-five percent of all monitored streams in the Shenandoah Case Study Area have a current risk of Elevated, Severe, or Acute. Of the 55 percent, 18 percent are chronically acidic today (US EPA, 2009, section 4.2.4.3).

Based on a deposition scenario for this study area that maintains current emission levels from 2020 to 2050, the simulation forecast indicates that a large number of streams would still have Elevated to Acute problems with acidity in 2050.

Biological effects of increased acidification documented in the Shenandoah Case Study Area include a decrease in the condition factor in blacknose dace and a decrease in fish biodiversity associated with decreasing stream ANC. On average, the fish species richness is

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lower by one fish species for every 21 μeq/L decrease in ANC in Shenandoah National Park streams (US EPA, 2008, section 3.2.3.4).

ii. Terrestrial acidification

The ISA identified a variety of indicators that can be used to measure the effects of acidification in soils. Most effects of terrestrial acidification are observed in sensitive forest ecosystem in the U.S. Tree health has been linked to the availability of base cations (BC) in soil (such as Ca^{2+}, Mg^{2+} and K^{+}), as well as soil aluminum (Al) content. Tree species show a range of sensitivities to Ca/Al and BC/Al soil molar ratios, therefore these are good chemical indicators because they directly relate to the biological effects. Critical BC/Al molar ratios for a large variety of tree species ranged from 0.2 to 0.8. This range is similar to critical ratios of Ca/Al. Plant toxicity or nutrient antagonism was reported to occur at Ca/Al molar ratios ranging from 0.2 to 2.5 (US EPA, 2009).

There has been no systematic national survey of terrestrial ecosystems to determine the extent and distribution of terrestrial ecosystem sensitivity to the effects of acidifying deposition. However, one preliminary national evaluation estimated that ~15 percent of forest ecosystems in the U.S. exceed the estimated critical load based on soil ANC leaching for sulfur and nitgogen deposition by >250 eq/ha/yr (McNulty et al., 2007). Forests of the Adirondack Mountains of New York, Green Mountains of Vermont, White Mountains of New Hampshire, the Allegheny Plateau of Pennsylvania and high-elevation forest ecosystems in the southern Appalachians are the regions most sensitive to terrestrial acidification effects from acidifying deposition (US EPA, 2008, section 3.2.4.2). While studies show some recovery of surface waters, there are widespread measurements of ongoing depletion of exchangeable base cations in forest soils in

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the northeastern U.S. despite recent decreases in acidifying deposition, indicating a slow recovery time.

In the REA, a critical load analysis was performed for sugar maple and red spruce forests in the eastern U.S. by using BC/Al ratio in acidified forest soils as an indicator to assess the impact of nitrogen and sulfur deposition on tree health. These are the two most commonly studied tree species in North America for effects of acidification. At a BC/Al ratio of 1.2, red spruce growth can be decreased by 20 percent. Sugar maple growth can be decreased by 20 percent at a BC/Al ratio of 0.6 (US EPA, 2009, section 4.4). The REA analysis determined the health of at least a portion of the sugar maple and red spruce growing in the U.S. may have been compromised with acidifying total nitrogen and sulfur deposition. Specifically, total nitrogen and sulfur deposition levels exceeded three selected critical loads for tree growth in 3 percent to 75 percent of all sugar maple plots across 24 states -- that is, it exceeded the highest (least stringent) of the three critical loads in 3 percent of plots, and the lowest (most stringent) in 75 percent of plots. For red spruce, total nitrogen and sulfur deposition levels exceeded three selected critical loads in 3 percent to 36 percent of all red spruce plots across eight states (US EPA, 2009, section 4.4).

c. Key uncertainties associated with acidification

There are different levels of uncertainty associated with relationships between deposition, ecological effects and ecological indicators. In Chapter 7 of the REA, the case study analyses associated with each targeted effect area were synthesized by identifying the strengths, limitations, and uncertainties associated with the available data, modeling approach, and relationship between the selected ecological indicator and atmospheric deposition as described by the ecological effect function (US EPA, 2009, Figure 1-1). A further discussion of

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uncertainty in aquatic and terrestrial ecosystems is presented below. The key uncertainties were characterized as follows to evaluate the strength of the scientific basis for setting a national standard to protect against a given effect (US EPA, 2009, section 7):

(1) Data Availability: high, medium or low quality. This criterion is based on the availability and robustness of data sets, monitoring networks, availability of data that allows for extrapolation to larger assessment areas and input parameters for modeling and developing the ecological effect function. The scientific basis for the ecological indicator selected is also incorporated into this criterion.

(2) Modeling Approach: high, fairly high, intermediate, or low confidence. This value is based on the strengths and limitations of the models used in the analysis and how accepted they are by the scientific community for their application in this analysis.

(3) Ecological Effect Function: high, fairly high, intermediate or low confidence. This ranking is based on how well the ecological effect function describes the relationship between atmospheric deposition and the ecological indicator of an effect.

i. Aquatic acidification

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

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There is fairly high confidence associated with modeling and input parameters. Uncertainties in water quality estimates (i.e., ANC) from MAGIC were derived from multiple site calibrations. Pre-acidification refers to retrospective modeling to estimate water quality conditions before man-made contributions of acidifying inputs. The models are evaluated under current conditions to determine how well they replicate observed ANC values. The 95 percent confidence interval for pre-acidification of lakes was an average of 15 μeq/L difference in ANC concentrations, or 10 percent, and 8 μeq/L, or 5 percent, for streams (US EPA, 2009, section 7.1.2). The use of the critical load model to estimate aquatic critical loads is limited by the uncertainties associated with runoff and surface water measurements and in estimating the catchment supply of base cations from the weathering of bedrock and soils (McNulty et al., 2007).

ii. Terrestrial acidification

The available data used to quantify the targeted effect of terrestrial acidification are robust and considered high quality. The U.S. Forest Service-Kane Experimental Forest and significant amounts of research work in the Allegheny Plateau have produced extensive, peer-reviewed data sets. Sugar maple and red spruce were the focus of the REA since they are demonstrated to be negatively affected by soil available Ca^{2+} depletion and high concentrations of available Al, and occur in areas that receive high acidifying deposition. There is high confidence about the use of the REA terrestrial acidification data and their value for extrapolating to a larger regional population of forests.

There is high confidence associated with the models, input parameters, and assessment of uncertainty used in the case study for terrestrial acidification. The Simple Mass Balance (SMB) model, a commonly used and widely applied approach for estimating critical loads, was used in

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the REA analysis (US EPA, 2008, section 7.2.2). There is fairly high confidence associated with the ecological effect function developed for terrestrial acidification (US EPA, 2009, section 7.2.3).

3. *Nutrient Enrichment Effects Associated with Deposition of Oxides of Nitrogen*

The following summary is a concise overview of the known or anticipated effects caused by nitrogen nutrient enrichment to ecosystems within the United States. Nutrient-enrichment affects terrestrial, freshwater and estuarine ecosystems. Nitrogen deposition is a major source of anthropogenic nitrogen. For many terrestrial and freshwater ecosystems other sources of nitrogen including fertilizer and waste treatment are greater than deposition. Nitrogen deposition often contributes to nitrogen-enrichment effects in estuaries, but does not drive the effects since other sources of nitrogen greatly exceed nitrogen deposition. Both oxides of nitrogen and NH_x contribute to nitrogen deposition. For the most part, nitrogen effects on ecosystems do not depend on whether the nitrogen is in oxidized or reduced form. Thus, this summary focuses on the effects of nitrogen deposition in total.

a. Nature of nutrient enrichment-related ecosystem responses

The ISA found that deposition of nitrogen, including oxides of nitrogen and NH_x, leads to the nitrogen enrichment of ecosystems (US EPA 2008). In the process of nitrogen enrichment, biogeochemical components of terrestrial and freshwater aquatic ecosystems are altered in a way that leads to effects on biological organisms.

i. Aquatic ecosystems

In freshwater ecosystems, the evidence is sufficient to infer a causal relationship between nitrogen deposition and the alteration of biogeochemical cycling in freshwater aquatic ecosystems (US EPA, 2008, section 3.3.2.3). Nitrogen deposition is the main source of nitrogen

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enrichment to headwater streams, lower order streams and high elevation lakes. The most common chemical indicators that were studied included NO_3^- and dissolved inorganic nitrogen (DIN) concentration in surface waters as well as the ratio of chlorophyll *a* to total phosphorus. Elevated surface water NO_3^- concentrations occur in both the eastern and western U.S. Studies report a significant correlation between nitrogen deposition and lake biogeochemistry by identifying a correlation between wet deposition and DIN and the ratio of chlorophyll *a* to total phosphate. Recent evidence provides examples of lakes and streams that are limited by nitrogen and show signs of eutrophication in response to nitrogen addition.

The evidence is sufficient to infer a causal relationship between nitrogen deposition and the alteration of species richness, species composition and biodiversity in freshwater aquatic ecosystems (US EPA, 2008, section 3.3.5.3). Increased nitrogen deposition can cause a shift in community composition and reduce algal biodiversity, especially in sensitive oligotrophic lakes.

In the ISA, the evidence is sufficient to infer a causal relationship between nitrogen deposition and the biogeochemical cycling of nitrogen and carbon in estuaries (US EPA, 2008, section 4.3.4.1 and 3.3.2.3). In general, estuaries tend to be nitrogen-limited, and many currently receive high levels of nitrogen input from human activities (US EPA, 2009, section 5.1.1). It is unknown if atmospheric deposition alone is sufficient to cause eutrophication; however, the contribution of atmospheric nitrogen deposition to total nitrogen load is calculated for some estuaries and can be >40 percent (US EPA, 2009, section 5.1.1).

The evidence is sufficient to infer a causal relationship between nitrogen deposition and the alteration of species richness, species composition and biodiversity in estuarine ecosystems (US EPA, 2008, section 4.3.4.2 and 3.3.5.4). Atmospheric and non-atmospheric sources of nitrogen contribute to increased phytoplankton and algal productivity, leading to eutrophication.

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Shifts in community composition, reduced hypolimnetic dissolved oxygen (DO), decreases in biodiversity, and mortality of submerged aquatic vegetation are associated with increased N deposition in estuarine systems.

ii. Terrestrial Ecosystems

The evidence is sufficient to infer a causal relationship between nitrogen deposition and the alteration of biogeochemical cycling in terrestrial ecosystems (US EPA, 2008, section 4.3.1.1 and 3.3.2.1). This is supported by numerous observational, deposition gradient and field addition experiments in sensitive ecosystems. The leaching of NO_3- in soil drainage waters and the export of NO_3- in stream water were identified as two of the primary indictors of nitrogen enrichment. Several nitrogen-addition studies indicate that NO_3- leaching is induced by chronic additions of nitrogen. Studies identified in the ISA found that surface water NO_3- concentrations exceeded 1 μeq/L in watersheds receiving about 9 to 13 kg N/ha/yr of atmospheric nitrogen deposition. Nitrogen deposition disrupts the nutrient balance of ecosystems with numerous biogeochemical effects. . The chemical indicators that are typically measured include NO_3^- leaching, soil C:N ratio, rates of nitrogen mineralization, nitrification, denitrification, foliar nitrogen concentration, and soil water NO_3 − and NH_4^+ concentrations. Note that nitrogen saturation (nitrogen leaching from ecosystems) does not need to occur to cause effects. Substantial leaching of NO_3− from forest soils to stream water can acidify downstream waters, leading to effects described in the previous section on aquatic acidification. Due to the complexity of interactions between the nitrogen and carbon cycling, the effects of nitrogen on carbon budgets (quantified input and output of carbon to the ecosystem) are variable. Regional trends in net ecosystem productivity (NEP) of forests (not managed for silviculture) have been estimated through models based on gradient studies and meta-analysis. Atmospheric

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nitrogen deposition has been shown to cause increased litter accumulation and carbon storage in above-ground woody biomass. In the West, this has lead to increased susceptibility to more severe fires. Less is known regarding the effects of nitrogen deposition on carbon budgets of non-forest ecosystems.

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

iii. *Ecosystem sensitivity to nutrient enrichment*

The numerous ecosystem types that occur across the U.S. have a broad range of sensitivity to nitrogen deposition (US EPA, 2008, Table 4-4). Increased deposition to nitrogen-limited ecosystems can lead to production increases that may be either beneficial or adverse depending on the system and management goals.

Organisms in their natural environment are commonly adapted to a specific regime of nutrient availability. Change in the availability of one important nutrient, such as nitrogen, may result in an imbalance in ecological stoichiometry, with effects on ecosystem processes, structure

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and function. In general, nitrogen deposition to terrestrial ecosystems causes accelerated growth rates in some species deemed desirable in commercial forests but may lead to altered competitive interactions among species and nutrient imbalances, ultimately affecting biodiversity. The onset of these effects occurs with nitrogen deposition levels as low as 3 kg N/ha/yr in sensitive terrestrial ecosystems to nitrogen deposition. In aquatic ecosystems, nitrogen that is both leached from the soil and directly deposited to the water surface can pollute the surface water. This causes alteration of the diatom community at levels as low as 1.5 kg N/ha/yr in sensitive freshwater ecosystems.

The degree of ecosystem effects lies at the intersection of nitrogen loading and nitrogen-sensitivity. Nitrogen-sensitivity is predominately driven by the degree to which growth is limited by nitrogen availability. Grasslands in the western U.S. are typically nitrogen-limited ecosystems dominated by a diverse mix of perennial forbs and grass species. A meta-analysis discussed in the ISA (US EPA, 2008, section 3.3.3), indicated that nitrogen fertilization increased aboveground growth in all non-forest ecosystems except for deserts. In other words, almost all terrestrial ecosystems are nitrogen-limited and will be altered by the addition of anthropogenic nitrogen. Likewise, a freshwater lake or stream must be nitrogen-limited to be sensitive to nitrogen-mediated eutrophication. There are many examples of fresh waters that are nitrogen-limited or nitrogen and phosphorous (P) co-limited (US EPA, 2008, section 3.3.3.2). A large dataset meta-analysis discussed in the ISA (US EPA, 2008, section 3.3.3.2), found that nitrogen-limitation occurred as frequently as phosphorous-limitation in freshwater ecosystems. Additional factors that govern the sensitivity of ecosystems to nutrient enrichment from nitrogen deposition include rates and form of nitrogen deposition, elevation, climate, species composition, plant growth rate, length of growing season, and soil nitrogen retention capacity (US EPA, 2008,

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section 4.3). Less is known about the extent and distribution of the terrestrial ecosystems in the U.S. that are most sensitive to the effects of nutrient enrichment from atmospheric nirogen deposition compared to acidification.

Because the productivity of estuarine and near shore marine ecosystems is generally limited by the availability of nitrogen, they are susceptible to the eutrophication effect of nitrogen deposition (US EPA, 2008, section 4.3.4.1). A recent national assessment of eutrophic conditions in estuaries found the most eutrophic estuaries were generally those that had large watershed-to-estuarine surface area, high human population density, high rainfall and runoff, low dilution and low flushing rates. In the REA, the National Oceanic and Atmospheric Administration's (NOAA) National Estuarine Eutrophication Assessment (NEEA) assessment tool, Assessment of Estuarine Tropic Status (ASSETS) categorical Eutrophication Index (EI) was used to evaluate eutrophication due to atmospheric loading of nitrogen. The ASSETS EI is an estimation of the likelihood that an estuary is experiencing eutrophication or will experience eutrophication based on five ecological indicators: chlorophyll *a*, macroalgae, dissolved oxygen, nuisance/toxic algal blooms and submerged aquatic vegetation (SAV).

In the REA, two regions were selected for case study analysis using ASSETS EI, the Chesapeake Bay and Pamlico Sound. Both regions received an ASSETS EI rating of Bad indicating that the estuary had moderate to high pressure due to overall human influence and a moderate high to high eutrophic condition (US EPA, 2009, sections 5.2.4.1 and 5.2.4.2). These results were then considered with SPAtially Referenced Regression on Watershed Attributes (SPARROW) modeling to develop a response curve to examine the role of atmospheric nitrogen deposition in achieving a desired decrease in load. To change the Neuse River Estuary's EI score from Bad to Poor not only must 100 percent of the total atmospheric nitrogen deposition be

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eliminated, but considerably more nitrogen from other sources as well must be controlled (US EPA, 2009, section 5.2.7.2). In the Potomac River estuary, a 78 percent decrease of total nitrogen could move the EI score from Bad to Poor (US EPA, 2009, section 5.2.7.1). The results of this analysis indicated decreases in atmospheric deposition alone could not eliminate coastal eutrophication problems due to multiple non-atmospheric nitrogen inputs (US EPA, 2009, section 7.3.3). However, the somewhat arbitrary discreteness of the EI scale can mask the benefits of decreases in nitrogen between categories.

In general, estuaries tend to be nitrogen-limited, and many currently receive high levels of nitrogen input from human activities to cause eutrophication. As reported in the ISA (US EPA, 2008, section 3.2.2.2), atmospheric nitrogen loads to estuaries in the U.S. are estimated to range from 2 to 8 percent for Guadalupe Bay, Texas on the lowest end to as high as 72 percent for St. Catherines-Sapelo estuary, Georgia. The Chesapeake Bay is an example of a large, well-studied and severely eutrophic estuary that is calculated to receive as much as 30 percent of its total nitrogen load from the atmosphere.

b. Magnitude of ecosystem responses

i. Aquatic ecosystems

The magnitude of ecosystem response may be thought of on two time scales, current conditions and how ecosystems have been altered since the onset of anthropogenic nitrogen deposition. As noted previously, studies found that nitrogen-limitation occurs as frequently as phosphorous-limitation in freshwater ecosystems (US EPA, 2008, section 3.3.3.2). Recently, a comprehensive study of available data from the northern hemisphere surveys of lakes along gradients of nitrogen deposition show increased inorganic nitrogen concentration and productivity to be correlated with atmospheric nitrogen deposition. The results are unequivocal

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evidence of nitrogen limitation in lakes with low ambient inputs of nitrogen, and increased nitrogen concentrations in lakes receiving nitrogen solely from atmospheric nitrogen deposition. It has been suggested that most lakes in the northern hemisphere may have originally been nitrogen-limited, and that atmospheric nitrogen deposition has changed the balance of nitrogen and phosphorous in lakes.

Available data suggest that the increases in total nitrogen deposition do not have to be large to elicit an ecological effect. For example, a hindcasting exercise determined that the change in Rocky Mountain National Park lake algae that occurred between 1850 and 1964 was associated with an increase in wet nitrogen deposition that was only about 1.5 kg N/ha. Similar changes inferred from lake sediment cores of the Beartooth Mountains of Wyoming also occurred at about 1.5 kg N/ha deposition. Pre-industrial inorganic nitrogen deposition is estimated to have been only 0.1 to 0.7 kg N/ha based on measurements from remote parts of the world. In the western U.S., pre-industrial, or background, inorganic nitrogen deposition was estimated by to range from 0.4 to 0.7 kg N/ha/yr.

Eutrophication effects from nitrogen deposition are most likely to be manifested in undisturbed, low nutrient surface waters such as those found in the higher elevation areas of the western U.S. The most severe eutrophication from nitrogen deposition effects is expected downwind of major urban and agricultural centers. High concentrations of lake or streamwater NO_3^-, indicative of ecosystem saturation, have been found at a variety of locations throughout the U.S., including the San Bernardino and San Gabriel Mountains within the Los Angeles Air Basin, the Front Range of Colorado, the Allegheny mountains of West Virginia, the Catskill Mountains of New York, the Adirondack Mountains of New York, and the Great Smoky Mountains in Tennessee (US EPA, 2008, section 3.3.8).

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

There is a scientific consensus (US EPA, 2008, section 4.3.4) that nitrogen-driven eutrophication in shallow estuaries has increased over the past several decades and that the environmental degradation of coastal ecosystems due to nitrogen, phosphorus, and other inputs is now a widespread occurrence. For example, the frequency of phytoplankton blooms and the extent and severity of hypoxia have increased in the Chesapeake Bay and Pamlico estuaries in North Carolina and along the continental shelf adjacent to the Mississippi and Atchafalaya rivers' discharges to the Gulf of Mexico.

A recent national assessment of eutrophic conditions in estuaries found that 65 percent of the assessed systems had moderate to high overall eutrophic conditions Most eutrophic estuaries occurred in the mid-Atlantic region and the estuaries with the lowest degree of eutrophication were in the North Atlantic. Other regions had mixtures of low, moderate, and high degrees of eutrophication (US EPA, 2008, section 4.3.4.3).

The mid-Atlantic region is the most heavily impacted area in terms of moderate or high loss of submerged aquatic vegetation due to eutrophication (US EPA, 2008, section 4.3.4.2). Submerged aquatic vegetation is important to the quality of estuarine ecosystem habitats because

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it provides habitat for a variety of aquatic organisms, absorbs excess nutrients, and traps sediments (US EPA, 2008, section 4.3.4.2). It is partly because many estuaries and near-coastal marine waters are degraded by nutrient enrichment that they are highly sensitive to potential negative impacts from nitrogen addition from atmospheric deposition.

ii. Terrestrial ecosystems

Little is known about the full extent and distribution of the terrestrial ecosystems in the U.S. that are most sensitive to impacts caused by nutrient enrichment from atmospheric nitrogen deposition. As previously stated, most terrestrial ecosystems are nitrogen-limited, therefore they are sensitive to perturbation caused by nitrogen additions (US EPA, 2008, section 4.3.1). Effects are most likely to occur where areas of relatively high atmospheric N deposition intersect with nitrogen-limited plant communities. The alpine ecosystems of the Colorado Front Range, chaparral watersheds of the Sierra Nevada, lichen and vascular plant communities in the San Bernardino Mountains and the Pacific Northwest, and the southern California coastal sage scrub (CSS) community are among the most sensitive terrestrial ecosystems. There is growing evidence (US EPA, 2008, section 4.3.1.2) that existing grassland ecosystems in the western U.S. are being altered by elevated levels of N inputs, including inputs from atmospheric deposition.

In the eastern U.S., the degree of nitrogen saturation of the terrestrial ecosystem is often assessed in terms of the degree of NO_3^- leaching from watershed soils into ground water or surface water. Studies have estimated the number of surface waters at different stages of saturation across several regions in the eastern U.S. Of the 85 northeastern watersheds examined 60 percent were in Stage 1 or Stage 2 of nitrogen saturation on a scale of 0 (background or pretreatment) to 3 (visible decline). Of the northeastern sites for which adequate data were available for assessment, those in Stage 1 or 2 were most prevalent in the Adirondack and

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Catskill Mountains. Effects on individual plant species have not been well studied in the U.S. More is known about the sensitivity of particular plant communities. Based largely on results obtained in more extensive studies conducted in Europe, it is expected that the more sensitive terrestrial ecosystems include hardwood forests, alpine meadows, arid and semi-arid lands, and grassland ecosystems (US EPA, 2008, section 3.3.5).

The REA used published research results (US EPA, 2009, section 5.3.1 and US EPA, 2008, Table 4.4) to identify meaningful ecological benchmarks associated with different levels of atmospheric nitrogen deposition. These are illustrated in Figure 3-4 of the PA. The sensitive areas and ecological indicators identified by the ISA were analyzed further in the REA to create a national map that illustrates effects observed from ambient and experimental atmospheric nitrogen deposition loads in relation to Community Multi-scale Air Quality (CMAQ) 2002 modeling results and National Atmospheric Deposition Program (NADP) monitoring data. This map, reproduced in Figure 3-5 of the PA, depicts the sites where empirical effects of terrestrial nutrient enrichment have been observed and site proximity to elevated atmospheric nitrogen deposition.

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

The ecological benchmarks that were identified for the CSS and the MCF communities are included in the suite of benchmarks identified in the ISA (US EPA, 2008, section 3.3). There

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are sufficient data to confidently relate the ecological effect to a loading of atmospheric nitrogen. For the CSS community, the following ecological benchmarks were identified:

(1) 3.3 kg N/ha/yr – the amount of nitrogen uptake by a vigorous stand of CSS; above this level, nitrogen may no longer be limiting

(2) 10 kg N/ha/yr – mycorrhizal community changes

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

(1) 3.1 kg N/ha/yr – shift from sensitive to tolerant lichen species

(2) 5.2 kg N/ha/yr – dominance of the tolerant lichen species

(3) 10.2 kg N/ha/yr – loss of sensitive lichen species

(4) 17 kg N/ha/yr – leaching of nitrate into streams.

These benchmarks, ranging from 3.1 to 17 kg N/ha/yr, were compared to 2002 CMAQ/NADP data to discern any associations between atmospheric deposition and changing communities. Evidence supports the finding that nitrogen alters CSS and MCF communities. Key findings include the following: 2002 CMAQ/NADP nitrogen deposition data show that the 3.3 kg N/ha/yr benchmark has been exceeded in more than 93 percent of CSS areas (654,048 ha). These deposition levels are a driving force in the degradation of CSS communities. Although CSS decline has been observed in the absence of fire, the contributions of deposition and fire to the CSS decline require further research. The CSS is fragmented into many small parcels, and the 2002 CMAQ/NADP 12-km grid data are not fine enough to fully validate the relationship between CSS distribution, nitrogen deposition, and fire. The 2002 CMAQ/NADP nitrogen deposition data exceeds the 3.1 kg N/ha/yr benchmark in more than 38 percent (1,099,133 ha) of MCF areas, and nitrate leaching has been observed in surface waters. Ozone effects confound

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nitrogen effects on MCF acidophyte lichen, and the interrelationship between fire and nitrogen cycling requires additional research.

c. Key uncertainties associated with nutrient enrichment

There are different levels of uncertainty associated with relationships between deposition, ecological effects and ecological indicators. The criteria used in the REA to evaluate the degree of confidence in the data, modeling and ecological effect function are detailed in chapter 7 of the REA. Below is a discussion of uncertainty relating aquatic and terrestrial ecosystems to nutrient enrichment effects.

i. Aquatic ecosystems

The approach for assessing atmospheric contributions to total nitrogen loading in the REA was to consider the main-stem river to an estuary (including the estuary) rather than an entire estuary system or bay. The biological indicators used in the NOAA ASSETS EI required the evaluation of many national databases including the US Geological Survey National Water Quality Assessment (NAWQA) files, EPA's STORage and RETrieval (STORET) database, NOAA's Estuarine Drainage Areas data and EPA's water quality standards nutrient criteria for rivers and lakes (US EPA, 2009, Appendix 6 and Table 1.2.-1). Both the SPARROW modeling for nitrogen loads and assessment of estuary conditions under NOAA ASSETS EI, have been applied on a national scale. The REA concludes that the available data are medium quality with intermediate confidence about the use of these data and their values for extrapolating to a larger regional area (US EPA, 2009, section 7.3.1). Intermediate confidence is associated with the modeling approach using ASSETS EI and SPARROW. The REA states there is low confidence with the ecological effect function due to the results of the analysis which indicated that

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reductions in atmospheric deposition alone could not solve coastal eutrophication problems due to multiple non-atmospheric nitrogen inputs (US EPA, 2009, section 7.3.3).

ii. *Terrestrial ecosystems*

Ecological thresholds are identified for CSS and MCF areas and these data are considered to be of high quality, however, the ability to extrapolate these data to larger regional areas is limited (US EPA, 2009, section 7.4.1). No quantitative modeling was conducted or ecological effect function developed for terrestrial nutrient enrichment reflecting the uncertainties associated with these depositional effects.

4. *Other Ecological Effects*

It is stated in the ISA (US EPA, 2008, section 3.4.1 and 4.5) that mercury is a highly neurotoxic contaminant that enters the food web as a methylated compound, methylmercury (MeHg). Mercury is principally methylated by sulfur-reducing bacteria and can be taken up by microorganisms, zooplankton and macroinvertebrates. The contaminant is concentrated in higher trophic levels, including fish eaten by humans. Experimental evidence has established that only inconsequential amounts of MeHg can be produced in the absence of sulfate. Once MdHg is present, other variables influence how much accumulates in fish, but elevated mercury levels in fish can only occur where substantial amounts of MeHg are present. Current evidence indicates that in watersheds where mercury is present, increased oxides of sulfur deposition very likely results in additional production of MeHg which leads to greater accumulation of MeHg concentrations in fish. With respect to sulfur deposition and mercury methylation, the final ISA determined that "[t]he evidence is sufficient to infer a causal relationship between sulfur deposition and increased mercury methylation in wetlands and aquatic environments."

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The production of meaningful amounts of MeHg requires the presence of SO_4^{2-} and mercury, and where mercury is present, increased availability of SO_4^{2-} results in increased production of MeHg. There is increasing evidence on the relationship between sulfur deposition and increased methylation of mercury in aquatic environments; this effect occurs only where other factors are present at levels within a range to allow methylation. The production of MeHg requires the presence of SO_4^{2-} and mercury, but the amount of MeHg produced varies with oxygen content, temperature, pH, and supply of labile organic carbon (US EPA, 2008, section 3.4). In watersheds where changes in sulfate deposition did not produce an effect, one or several of those interacting factors were not in the range required for meaningful methylation to occur (US EPA, 2008, section 3.4). Watersheds with conditions known to be conducive to mercury methylation can be found in the northeastern U.S. and southeastern Canada.

While the relationship between sulfur and MeHg production was concluded to be causal in the ISA, the REA concluded that there was insufficient evidence to quantify the relationship between sulfur and MeHg. Therefore only a qualitative assessment was included in chapter 6 of the REA. The PA was then unable to make a determination as to the adequacy of the existing SO_2 standards in protecting against welfare effects associated with increased mercury methylation.

B. Risk and Exposure Assessment

The risk and exposure assessment conducted for the current review was developed to describe potential risk from current and future deposition of oxides of nitrogen and sulfur to sensitive ecosystems. The case study analyses in the REA show that there is confidence that known or anticipated adverse ecological effects are occurring under current ambient loadings of nitrogen and sulfur in sensitive ecosystems across the U.S. An overview of the material covered

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in the REA, a summary of the key findings from the air quality analyses, acidification and nutrient enrichment case studies, and general conclusions from evaluating additional welfare effects, are presented below.

1. *Overview of the Risk and Exposure Assessment*

The REA evaluates the relationships between atmospheric concentrations, deposition, biologically relevant exposures, targeted ecosystem effects, and ecosystem services. To evaluate the nature and magnitude of adverse effects associated with deposition, the REA also examines various ways to quantify the relationships between air quality indicators, deposition of biologically available forms of nitrogen and sulfur, ecologically relevant indicators relating to deposition, exposure and effects on sensitive receptors, and related effects resulting in changes in ecosystem structure and services. The intent is to determine the exposure metrics that incorporate the temporal considerations (i.e., biologically relevant timescales), pathways, and ecologically relevant indicators necessary to determine the effects on these ecosystems. To the extent feasible, the REA evaluates the overall load to the system for nitrogen and sulfur, as well as the variability in ecosystem responses to these pollutants. It also evaluates the contributions of atmospherically deposited nitrogen and sulfur individually relative to the combined atmospheric loadings of both elements together.. Since oxidized nitrogen is the listed criteria pollutant (currently measured by the ambient air quality indicator NO_2) for the atmospheric contribution to total nitrogen, the REA examines the contribution of nitrogen oxides to total reactive nitrogen in the atmosphere, relative to the contributions of reduced forms of nitrogen (e.g., ammonia, ammonium), to ultimately assess how a meaningful secondary NAAQS might be structured.

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The REA focuses on ecosystem welfare effects that result from the deposition of total reactive nitrogen and sulfur. Because ecosystems are diverse in biota, climate, geochemistry, and hydrology, response to pollutant exposures can vary greatly between ecosystems. In addition, these diverse ecosystems are not distributed evenly across the United States. To target nitrogen and sulfur acidification and nitrogen and sulfur enrichment, the REA addresses four main targeted ecosystem effects on terrestrial and aquatic systems identified by the ISA (US EPA, 2008): aquatic acidification due to nitrogen and sulfur; terrestrial acidification due to nitrogen and sulfur; aquatic nutrient enrichment, including eutrophication; and terrestrial nutrient enrichment.

In addition to these four targeted ecosystem effects, the REA also qualitatively addresses the influence of sulfur oxides deposition on MeHg production; nitrous oxide (N_2O) effects on climate; nitrogen effects on primary productivity and biogenic greenhouse gas (GHG) fluxes; and phytotoxic effects on plants.

Because the targeted ecosystem effects outlined above are not evenly distributed across the U.S., the REA identified case studies for each targeted effects based on ecosystems identified as sensitive to nitrogen and/or sulfur deposition effects. Eight case study areas and two supplemental study areas (Rocky Mountain National Park and Little Rock Lake, Wisconsin) are summarized in the REA based on ecosystem characteristics, indicators, and ecosystem service information. Case studies selected for aquatic acidification effects were the Adirondack Mountains and Shenandoah National Park. Kane Experimental Forest in Pennsylvania and Hubbard Brook Experimental Forest in New Hampshire were selected as case studies for terrestrial acidification. Aquatic nutrient enrichment case study locations were selected in the Potomac River Basin upstream of Chesapeake Bay and the Neuse River Basin upstream of the

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Pamlico Sound in North Carolina. The CSS communities in southern California and the MCF communities in the San Bernardino and Sierra Nevada Mountains of California were selected as case studies for terrestrial nutrient enrichment. Two supplemental areas were also chosen, one in Rocky Mountain National Park for terrestrial nutrient enrichment and one in Little Rock Lake, Wisconsin for aquatic nutrient enrichment.

2. *Key findings*

In summary, based on case study analyses, the REA concludes that known or anticipated adverse ecological effects are occurring under current conditions and further concludes that these adverse effects continue into the future. Key findings from the air quality analyses, acidification and nutrient enrichment case studies, as well as general conclusions from evaluating additional welfare effects, are summarized below.

a. Air quality analyses

The air quality analyses in the REA encompass the current emissions sources of nitrogen and sulfur, as well as atmospheric concentrations, estimates of deposition of total nitrogen, policy-relevant background, and non-atmospheric loadings of nitrogen and sulfur to ecosystems, both nationwide and in the case study areas. Spatial fields of deposition were created using wet deposition measurements from the NADP National Trends Network and dry deposition predictions from the 2002 CMAQ model simulation. Some key conclusions from this analysis are:

(1) Total reactive nitrogen deposition and sulfur deposition are much greater in the East compared to most areas of the West.

(2) These regional differences in deposition correspond to the regional differences in oxides of nitrogen and SO_2 concentrations and emissions, which are also higher in the East. Oxides of

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nitrogen emissions are much greater and generally more widespread than NH_3 emissions nationwide; high NH_3 emissions tend to be more local (e.g., eastern North Carolina) or sub-regional (e.g., the upper Midwest and Plains states). The relative amounts of oxidized versus reduced nitrogen deposition are consistent with the relative amounts of oxides of nitrogen and NH_3 emissions. Oxidized nitrogen deposition exceeds reduced nitrogen deposition in most of the case study areas; the major exception being the Neuse River/Neuse River Estuary Case Study Area.

(3) Reduced nitrogen deposition exceeds oxidized nitrogen deposition in the vicinity of local sources of NH_3.

(4) There can be relatively large spatial variations in both total reactive nitrogen deposition and sulfur deposition within a case study area; this occurs particularly in those areas that contain or are near a high emissions source of oxides of nitrogen, NH_3 and/or SO_2.

(5) The seasonal patterns in deposition differ between the case study areas. For the case study areas in the East, the season with the greatest amounts of total reactive nitrogen deposition correspond to the season with the greatest amounts of sulfur deposition. Deposition peaks in spring in the Adirondack, Hubbard Brook Experimental Forest, and Kane Experimental Forest case study areas, and it peaks in summer in the Potomac River/Potomac Estuary, Shenandoah, and Neuse River/Neuse River Estuary case study areas. For the case study areas in the West, there is less consistency in the seasons with greatest total reactive nitrogen and sulfur deposition in a given area. In general, both nitrogen and/or sulfur deposition peaks in spring or summer. The exception to this is the Sierra Nevada Range portion of the MCF Case Study Area, in which sulfur deposition is greatest in winter.

b. *Deposition-related aquatic acidification*

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The role of aquatic acidification in two eastern United States areas—northeastern New York's Adirondack area and the Shenandoah area in Virginia—was analyzed in the REA to assess surface water trends in SO_4^{2-} and NO_3^- concentrations and ANC levels and to affirm the understanding that reductions in deposition could influence the risk of acidification. Monitoring data from the EPA-administered TIME)/ LTM programs and the EMAP were assessed for the years 1990 to 2006, and past, present and future water quality levels were estimated using both steady-state and dynamic biogeochemical models.

Although wet deposition rates for SO_2 and oxides of nitrogen in the Adirondack Case Study Area have reduced since the mid-1990s, current concentrations are still well above pre-acidification (1860) conditions. The MAGIC modeling predicts NO_3^- and SO_4^{2-} are 17- and 5-fold higher today, respectively. The estimated average ANC for 44 lakes in the Adirondack Case Study Area is 62.1 μeq/L (± 15.7 μeq/L); 78 percent of all monitored lakes in the Adirondack Case Study Area have a current risk of Elevated, Severe, or Acute. Of the 78 percent, 31 percent experience episodic acidification, and 18 percent are chronically acidic today.

(1) Based on the steady-state critical load model for the year 2002, 18 percent, 28 percent, 44 percent, and 58 percent of 169 modeled lakes received combined total sulfur and nitrogen deposition that exceeded critical loads corresponding to ANC limits of 0, 20, 50, and 100 μeq/L respectively.

(2) Based on a deposition scenario that maintains current emission levels to 2020 and 2050, the simulation forecast indicates no improvement in water quality in the Adirondack Case Study Area. The percentage of lakes within the Elevated to Acute Concern classes remains the same in 2020 and 2050.

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(3) Since the mid-1990s, streams in the Shenandoah Case Study Area have shown slight declines in NO_3 and SO_4^{2-} concentrations in surface waters. The ANC levels increased from about 50 μeq/L in the early 1990s to >75 μeq/L until 2002, when ANC levels declined back to 1991–1992 levels. Current concentrations are still above pre-acidification (1860) conditions. The MAGIC modeling predicts surface water concentrations of NO_3 and SO_4^{2-} are 10- and 32-fold higher today, respectively. The estimated average ANC for 60 streams in the Shenandoah Case Study Area is 57.9 μeq/L (± 4.5 μeq/L). Fifty-five percent of all monitored streams in the Shenandoah Case Study Area have a current risk of Elevated, Severe, or Acute. Of the 55 percent, 18 percent experience episodic acidification, and 18 percent are chronically acidic today.

(4) Based on the steady-state critical load model for the year 2002, 52 percent, 72 percent, 85 percent and 93 percent of 60 modeled streams received combined total sulfur and nitrogen deposition that exceeded critical loads corresponding to ANC limits of 0, 20, 50, and 100 μeq/L respectively.

(5) Based on a deposition scenario that maintains current emission levels to 2020 and 2050, the simulation forecast indicates that a large number of streams would still have Elevated to Acute problems with acidity.

c. *Deposition-related terrestrial acidification*

The role of terrestrial acidification was examined in the REA using a critical load analysis for sugar maple and red spruce forests in the eastern U.S. by using the BC/Al ratio in acidified forest soils as an indicator to assess the impact of nitrogen and sulfur deposition on tree health. These are the two most commonly studied species in North America for impacts of acidification. At a BC/Al ratio of 1.2, red spruce growth can be reduced by 20 percent. Sugar

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maple growth can be reduced by 20 percent at a BC/Al ratio of 0.6. Key findings of the case study are summarized below.

(1) Case study results suggest that the health of at least a portion of the sugar maple and red spruce growing in the U.S. may have been compromised with acidifying total nitrogen and sulfur deposition in 2002. The 2002 CMAQ/NADP total nitrogen and sulfur deposition levels exceeded three selected critical loads in 3 percent to 75 percent of all sugar maple plots across 24 states. The three critical loads ranged from 6,008 to 107 eq/ha/yr for the BC/Al ratios of 0.6, 1.2, and 10.0 (increasing levels of tree protection). The 2002 CMAQ/NADP total nitrogen and sulfur deposition levels exceeded three selected critical loads in 3 percent to 36 percent of all red spruce plots across eight states. The three critical loads ranged from 4,278 to 180 eq/ha/yr for the Bc/Al ratios of 0.6, 1.2, and 10.0 (increasing levels of tree protection).

(2) The SMB model assumptions made for base cation weathering (Bcw) and forest soil ANC input parameters are the main sources of uncertainty since these parameters are rarely measured and require researchers to use default values.

(3) The pattern of case study results suggests that nitrogen and sulfur acidifying deposition in the sugar maple and red spruce forest areas studied were similar in magnitude to the critical loads for those areas and both ecosystems are likely to be sensitive to any future changes in the levels of deposition.

d. *Deposition-related aquatic nutrient enrichment*

The role of nitrogen deposition in two main stem rivers feeding their respective estuaries was analyzed in the REA to determine if decreases in deposition could influence the risk of eutrophication as predicted using the ASSETS EI scoring system in tandem with SPARROW

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modeling. This modeling approach provides a transferrable, intermediate-level analysis of the linkages between atmospheric deposition and receiving waters, while providing results on which conclusions could be drawn. A summary of findings follows:

(1) The 2002 CMAQ/NADP results showed that an estimated 40,770,000 kilograms (kg) of total nitrogen was deposited in the Potomac River watershed. The SPARROW modeling predicted that 7,380,000 kg N/yr of the deposited nitrogen reached the estuary (20 percent of the total load to the estuary). The overall ASSETS EI for the Potomac River and Potomac Estuary was Bad (based on all sources of N).

(2) To improve the Potomac River and Potomac Estuary ASSETS EI score from Bad to Poor, a decrease of at least 78 percent in the 2002 total nitrogen atmospheric deposition load to the watershed would be required.

(3) The 2002 CMAQ/NADP results showed that an estimated 18,340,000 kg of total nitrogen was deposited in the Neuse River watershed. The SPARROW modeling predicted that 1,150,000 kg N/yr of the deposited nitrogen reached the estuary (26 percent of the total load to the estuary). The overall ASSETS EI for the Neuse River/Neuse River Estuary was Bad.

(4) It was found that the Neuse River/Neuse River Estuary ASSETS EI score could not be improved from Bad to Poor with decreases only in the 2002 atmospheric deposition load to the watershed. Additional reductions would be required from other nitrogen sources within the watershed.

The small effect of decreasing atmospheric deposition in the Neuse River watershed is because the other nitrogen sources within the watershed are more influential than atmospheric deposition in affecting the total nitrogen loadings to the Neuse River Estuary, as estimated with the SPARROW model. A water body's response to nutrient loading depends on the magnitude

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(e.g., agricultural sources have a higher influence in the Neuse than in the Potomac), spatial distribution, and other characteristics of the sources within the watershed; therefore a reduction in nitrogen deposition does not always produce a linear response in reduced load to the estuary, as demonstrated by these two case studies.

e. Deposition-related terrestrial nutrient enrichment

California CSS and MCF communities were the focus of the Terrestrial Nutrient Enrichment Case Studies of the REA. Geographic information systems analysis supported a qualitative review of past field research to identify ecological benchmarks associated with CSS and mycorrhizal communities, as well as MCF's nutrient-sensitive acidophyte lichen communities, fine-root biomass in Ponderosa pine and leached nitrate in receiving waters. These benchmarks, ranging from 3.1 to 17 kg N/ha/yr, were compared to 2002 CMAQ/NADP data to discern any associations between atmospheric deposition and changing communities. Evidence supports the finding that nitrogen alters CSS and MCF. Key findings include the following:

(1) The 2002 CMAQ/NADP nitrogen deposition data show that the 3.3 kg N/ha/yr benchmark has been exceeded in more than 93 percent of CSS areas (654,048 ha). This suggests that such deposition is a driving force in the degradation of CSS communities. One potentially confounding factor is the role of fire. Although CSS decline has been observed in the absence of fire, the contributions of deposition and fire to the CSS decline require further research. The CSS is fragmented into many small parcels, and the 2002 CMAQ/NADP 12-km grid data are not fine enough to fully validate the relationship between CSS distribution, nitrogen deposition, and fire.

(2) The 2002 CMAQ/NADP nitrogen deposition data exceeds the 3.1 kg N/ha/yr benchmark in more than 38% (1,099,133 ha) of MCF areas, and nitrate leaching has been observed in

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surface waters. Ozone effects confound nitrogen effects on MCF acidophyte lichen, and the interrelationship between fire and nitrogen cycling requires additional research.

f. *Additional effects*

Ecological effects have also been documented across the U.S. where elevated nitrogen deposition has been observed, including the eastern slope of the Rocky Mountains where shifts in dominant algal species in alpine lakes have occurred where wet nitrogen deposition was only about 1.5 kg N/ha/yr. High alpine terrestrial communities have a low capacity to sequester nitrogen deposition, and monitored deposition exceeding 3 to 4 kg N/ha/yr could lead to community-level changes in plant species, lichens and mycorrhizae.

Additional welfare effects are documented, but examined less extensively, in the REA. These effects include qualitative discussions related to visibility and materials damage, such as corrosion, erosion, and soiling of paint and buildings which are being addressed in the PM NAAQS review currently underway. A discussion of the causal relationship between sulfur deposition (as sulfate, SO_4^{2-}) and increased mercury methylation in wetlands and aquatic environments is also included in the REA. On this subject the REA concludes that decreases in SO_4^{2-} deposition will likely result in decreases in MeHg concentration; however, spatial and biogeochemical variations nationally hinder establishing large scale dose-response relationships.

Several additional issues concerning oxides of nitrogen were addressed in the REA. Consideration was also given to N_2O, a potent GHG. The REA concluded that it is most appropriate to analyze the role of N_2O in the context of all of the GHGs rather than as part of the REA for this review. The REA considered nitrogen deposition and its correlation with the rate of photosynthesis and net primary productivity. Nitrogen addition ranging from 15.4 to 300 kg N/ha/yr is documented as increasing wetland N_2O production by an average of 207 percent

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across all ecosystems. Nitrogen addition ranging from 30 to 240 kg N/ha/yr increased CH_4 emissions by 115 percent, averaged across all ecosystems, and methane uptake was reduced by 38 percent averaged across all ecosystems when nitrogen addition ranged from 10 to 560 kg N/ha/yr, but reductions were only significant for coniferous and deciduous forests. The heterogeneity of ecosystems across the U.S., however, introduces variations into dose-response relationships.

The phytotoxic effects of oxides of nitrogen and sulfur on vegetation were also briefly discussed in the REA which concluded that since a unique secondary NAAQS exists for SO_2, and concentrations of nitric oxide (NO), NO_2 and PAN are rarely high enough to have phytotoxic effects on vegetation, further assessment was not warranted at this time.

3. Conclusions on Effects

For aquatic and terrestrial acidification effects, a similar conceptual approach was used (critical loads) to evaluate the impacts of multiple pollutants on an ecological endpoint, whereas the approaches used for aquatic and terrestrial nutrient enrichment were fundamentally distinct. Although the ecological indicators for aquatic and terrestrial acidification (i.e., ANC and BC/Al) are very different, both ecological indicators are well-correlated with effects such as reduced biodiversity and growth. While aquatic acidification is clearly the targeted effect area with the highest level of confidence, the relationship between atmospheric deposition and an ecological indicator is also quite strong for terrestrial acidification. The main drawback with the understanding of terrestrial acidification is that the data are based on laboratory responses rather than field measurements. Other stressors that are present in the field but that are not present in the laboratory may confound this relationship.

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For nutrient enrichment effects, the REA utilized different types of indicators for aquatic and terrestrial effects to assess both the likelihood of adverse effects to ecosystems and the relationship between adverse effects and atmospheric sources of oxides of nitrogen. The ecological indicator chosen for aquatic nutrient enrichment, the ASSETS EI, seems to be inadequate to relate atmospheric deposition to the targeted ecological effect, likely due to the many other confounding factors. Further, there is far less confidence associated with the understanding of aquatic nutrient enrichment because of the large contributions from non-atmospheric sources of nitrogen and the influence of both oxidized and reduced forms of nitrogen, particularly in large watersheds and coastal areas. However, a strong relationship exists between atmospheric deposition of nitrogen and ecological effects in high alpine lakes in the Rocky Mountains because atmospheric deposition is the only source of nitrogen to these systems. There is also a strong weight-of-evidence regarding the relationships between ecological effects attributable to terrestrial nitrogen nutrient enrichment; however, ozone and climate change may be confounding factors. In addition, the response for other species or species in other regions of the U.S. has not been quantified.

C. Adversity of Effects to Public Welfare

Characterizing a known or anticipated adverse effect to public welfare is an important component of developing any secondary NAAQS. According to the CAA, welfare effects include: "effects on soils, water, crops, vegetation, manmade materials, animals, wildlife, weather, visibility, and climate, damage to and deterioration of property, and hazards to transportation, as well as effect on economic values and on personal comfort and well-being, whether caused by transformation, conversion, or combination with other air pollutants" (CAA,

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Section 302(h)). While the text above lists a number of welfare effects, these effects do not define public welfare in and of themselves.

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

Similar concepts were used in past reviews of secondary NAAQS for ozone and PM (relating to visibility), as well as in initial reviews of effects from lead deposition. Because oxides of nitrogen and sulfur are deposited from ambient sources into ecosystems where they affect changes to organisms, populations and ecosystems, the concept of adversity to public welfare as a result of alterations in structure and function of ecosystems is an appropriate consideration for this review.

Based on information provided in the PA, the following section discusses how ecological effects from deposition of oxides of nitrogen and sulfur relate to adversity to public welfare. In the PA, public welfare was discussed in terms of loss of ecosystem services (defined below), which in some cases can be monetized. Each of the four main effect areas (aquatic and terrestrial acidification and aquatic and terrestrial nutrient over-enrichment) are discussed including current ecological effects and associated ecosystem services.

1. Ecosystem Services

The PA defines ecosystem services as the benefits individuals and organizations obtain from ecosystems. Ecosystem services can be classified as provisioning (food and water),

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regulating (control of climate and disease), cultural (recreational, existence, spiritual, educational), and supporting (nutrient cycling). Conceptually, changes in ecosystem services may be used to aid in characterizing a known or anticipated adverse effect to public welfare. In the REA and PA ecosystem services are discussed as a method of assessing the magnitude and significance to the public of resources affected by ambient concentrations of oxides of nitrogen and sulfur and deposition in sensitive ecosystems.

The EPA has in previous NAAQS reviews defined ecological goods and services for the purposes of a Regulatory Impact Analysis as the "outputs of ecological functions or processes that directly or indirectly contribute to social welfare or have the potential to do so in the future. Some outputs may be bought and sold, but most are not marketed." It is especially important to acknowledge that it is difficult to measure and/or monetize the goods and services supplied by ecosystems. It can be informative in characterizing adversity to public welfare to attempt to place an economic valuation on the set of goods and services that have been identified with respect to a change in policy however it must be noted that this valuation will be incomplete and illustrative only.

Knowledge about the relationships linking ambient concentrations and ecosystem services is considered in the PA as one method by which to inform a policy judgment on a known or anticipated adverse public welfare effect. For example, a change in an ecosystem structure and process, such as foliar injury, would be classified as an ecological effect, with the associated changes in ecosystem services, such as primary productivity, food availability, forest products, and aesthetics (e.g., scenic viewing), classified as public welfare effects. Additionally, changes in biodiversity would be classified as an ecological effect, and the associated changes in

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ecosystem services—productivity, existence (nonuse) value, recreational viewing and aesthetics—would also be classified as public welfare effects.

As described in chapters 4 and 5 of the REA, case study analyses were performed that link deposition in sensitive ecosystems to changes in a given ecological indicator (e.g., for aquatic acidification, to changes in ANC) and then to changes in ecosystems. Appendix 8 of the REA links the changes in ecosystems to the services they provide (e.g., fish species richness and its influence on recreational fishing). To the extent possible for each targeted effect area, the REA linked ambient concentrations of nitrogen and sulfur (i.e., ambient air quality indicators) to deposition in sensitive ecosystems (i.e., exposure pathways), and then to system response as measured by a given ecological indicator (e.g., lake and stream acidification as measured by ANC). The ecological effect (e.g., changes in fish species richness) was then, where possible, associated with changes in ecosystem services and the corresponding public welfare effects (e.g., recreational fishing).

2. Effects on Ecosystem Services

The process used to link ecological indicators to ecosystem services is discussed extensively in appendix 8 of the REA. In brief, for each case study area assessed, the ecological indicators are linked to an ecological response that is subsequently linked to associated services to the extent possible. For example, in the case study for aquatic acidification the chosen ecological indicator is ANC which can be linked to the ecosystem service of recreational fishing. Although recreational fishing losses are the only service effects that can be independently quantified or monetized at this time, there are numerous other ecosystem services that may be related to the ecological effects of acidification.

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While aquatic acidification is the focus of this proposed standard, the other effect areas were also analyzed in the REA and these ecosystems are being harmed by nitrogen and sulfur deposition and will obtain some measure of protection with any decrease in that deposition regardless of the reason for the decrease. The following summarizes the current levels of specific ecosystem services for aquatic and terrestrial acidification and aquatic and terrestrial nutrient over-enrichment and attempts to quantify and when possible monetize the harm to public welfare, as represented by ecosystem services, due to nitrogen and sulfur deposition.

a. *Aquatic Acidification*

Acidification of aquatic ecosystems primarily affects the ecosystem services that are derived from the fish and other aquatic life found in surface waters. In the northeastern United States, the surface waters affected by acidification are not a major source of commercially raised or caught fish; however, they are a source of food for some recreational and subsistence fishers and for other consumers. Although data and models are available for examining the effects on recreational fishing, relatively little data are available for measuring the effects on subsistence and other consumers. Inland waters also provide aesthetic and educational services along with non-use services, such as existence value (protection and preservation with no expectation of direct use). In general, inland surface waters such as lakes, rivers, and streams also provide a number of regulating services, playing a role in hydrological regimes and climate regulation. There is little evidence that acidification of freshwaters in the northeastern U.S. has significantly degraded these specific services; however, freshwater ecosystems also provide biological control services by providing environments that sustain delicate aquatic food chains. The toxic effects of acidification on fish and other aquatic life impair these services by disrupting the trophic structure of surface waters. Although it is difficult to quantify these services and how they are

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affected by acidification, it is worth noting that some of these services may be captured through measures of provisioning and cultural services. For example, these biological control services may serve as "intermediate" inputs that support the production of "final" recreational fishing and other cultural services.

As summarized in Chapter 4 of the PA, recent studies indicate that acidification of lakes and streams can result in significant loss in economic value. For example, data indicate that more than 9 percent of adults in the northeastern part of the country participate annually in freshwater fishing yielding 140 million freshwater fishing days. Each fishing day has an estimated average value per day of $35. Therefore, the implied total annual value of freshwater fishing in the northeastern U.S. was $5 billion in 2006. Embedded in these numbers is a degree of harm to recreational fishing services due to acidification that has occurred over time. These harms have not been quantified on a regional scale; however, a case study was conducted in the Adirondacks area (US EPA, 2011, section 4.4.2).

In the Adirondacks case study, estimates of changes in recreational fishing services were determined, as well as changes more broadly in "cultural" ecosystem services (including recreational, aesthetic, and nonuse services). First, the MAGIC model (US EPA, 2009, Appendix 8 and section 2.2) was applied to 44 lakes to predict what ANC levels would be under both "business as usual" conditions (i.e., allowing for some decline in deposition due to existing regulations) and pre-emission (i.e., background) conditions. Second, to estimate the recreational fishing impacts of aquatic acidification in these lakes, an existing model of recreational fishing demand and site choice was applied. This model predicts how recreational fishing patterns in the Adirondacks would differ and how much higher the average annual value of recreational fishing services would be for New York residents if lake ANC levels corresponded to background

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(rather than business as usual) conditions. To estimate impacts on a broader category of cultural (and some provisioning) ecosystem services, results from the Banzhaf et al (2006) valuation survey of New York residents were adapted and applied to this context. The survey used a contingent valuation approach to estimate the average annual household willingness to pay (WTP) for future reductions in the percent of Adirondack lakes impaired by acidification. The focus of the survey was on impacts on aquatic resources. Pretesting of the survey indicated that respondents nonetheless tended to assume that benefits would occur in the condition of birds and forests as well as in recreational fishing.

By extrapolating the 44 lake Adirondack case study to all 3,000 Adirondack lakes and by applying the WTP survey results to all New York residents, the study estimated aggregated benefits between $300 and $800 million annually for the equivalent of improving lakes in the Adirondacks region to an ANC level of 50 μeq/L. The REA estimated 44 percent of the Adirondack lakes currently fall below an ANC of 50 μeq/L. Several states have set goals for improving the acid status of lakes and streams, generally targeting ANC in the range of 50 to 60 μeq/L, and have engaged in costly activities to decrease acidification.

These results imply significant value to the public in addition to those derived from recreational fishing services. Note that the results are only applicable to improvements in the Adirondacks valued by residents of New York. If similar benefits exist in other acid-impacted areas, benefits for the nation as a whole could be substantial. The analysis provides results on only a subset of the impacts of acidification on ecosystem services and suggests that the overall impact on these services is likely to be substantial.

b. Terrestrial Acidification

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Chapters 4.4.3 and 4.4.4 of the PA review several economic studies of areas sensitive to terrestrial acidification. Forests in the northeastern U.S. provide several important and valuable provisioning ecosystem services, which are reflected in the production and sales of tree products. Sugar maples are a particularly important commercial hardwood tree species in the United States, producing timber and maple syrup that provide hundreds of millions of dollars in economic value annually. Red spruce is also used in a variety of wood products and provides up to $100 million in economic value annually. Although the data do not exist to directly link acidification damages to economic values of lost recreational ecosystem services in forests, these resources are valuable to the public. A recent study, reviewed in the PA, suggests that the total annual value of recreational off-road driving was more than $9 billion and the value of hunting and wildlife viewing was more than $4 billion each in the northeastern States. The EPA is not able to quantify at this time the specific effects on these values of acid deposition, or of any specific reductions in deposition, relative to the effects of many other factors that may affect them.

c. Nutrient Enrichment

Chapters 4.4.5 and 4.4.6 of the PA summarize economic studies of east coast estuaries affected by nutrient over-enrichment or eutrophication. Estuaries in the eastern United States are important for fish and shellfish production. The estuaries are capable of supporting large stocks of resident commercial species, and they serve as the breeding grounds and interim habitat for several migratory species. To provide an indication of the magnitude of provisioning services associated with coastal fisheries, from 2005 to 2007, the average value of total catch was $1.5 billion per year in 15 East Coast states. Estuaries also provide an important and substantial variety of cultural ecosystem services, including water-based recreational and aesthetic services.

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For example, data indicate that 4.8 percent of the population in coastal states from North Carolina to Massachusetts participated in saltwater fishing, with a total of 26 million saltwater fishing days in 2006. Based on estimates in the PA, total recreational value from these saltwater fishing days was approximately $1.3 billion. Recreational participation estimates for 1999–2000 showed almost 6 million individuals participated in motorboating in coastal states from North Carolina to Massachusetts. The aggregate value of these coastal motorboating outings was $2 billion per year. EPA is not able to quantify at this time the specific effects on these values of nitrogen deposition, or of any specific reductions in deposition, relative to the effects of many other factors that may affect them.

Terrestrial ecosystems can also suffer from nutrient over-enrichment. Each ecosystem is different in its composition of species and nutrient requirements. Changes to individual ecosystems from changes in nitrogen deposition can be hard to assess economically. Relative recreational values are often determined by public use information. Chapter 4.4.7 of the PA reviewed studies related to park use in California. Data from California State Parks indicate that in 2002, 68.7 percent of adult residents participated in trail hiking for an average of 24.1 days per year. The analyses in the PA indicate that the aggregate annual benefit for California residents from trail hiking in 2007 was $11.59 billion. EPA is not able to quantify at this time the specific effects on these values of nitrogen deposition, or of any specific reductions in deposition, relative to the effects of many other factors that may affect them.

The PA also identified fire regulation as a service that could be affected by nutrient over-enrichment of the CSS and MCF ecosystems by encouraging growth of more flammable grasses, increasing fuel loads, and altering the fire cycle. Over the 5-year period from 2004 to 2008, Southern California experienced, on average, over 4,000 fires per year, burning, on average, over

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400,000 acres per year. It is not possible at this time to quantify the contribution of nitrogen deposition, among many other factors, to increased fire risk.

3. Summary

Adversity to public welfare can be understood by looking at how deposition of oxides of nitrogen and sulfur affect the ecological functions of an ecosystem (see II.A.), and then understanding the ecosystem services that are degraded. The monetized value of the ecosystem services provided by ecosystems that are sensitive to deposition of oxides of nitrogen and sulfur are in the billions of dollars each year, though it is not possible to quantify or monetize at this time the effects on these values of nitrogen and sulfur deposition or of any changes in deposition that may result from new secondary standards. Many lakes and streams are known to be degraded by acidic deposition which affects recreational fishing and tourism. Forest growth is likely suffering from acidic deposition in sensitive areas affecting red spruce and sugar maple timber production, sugar maple syrup production, hiking, aesthetic enjoyment and tourism. Nitrogen deposition contributes significantly to eutrophication in many estuaries affecting fish production, swimming, boating, aesthetic enjoyment and tourism. Ecosystem services are likely affected by nutrient enrichment in many natural and scenic terrestrial areas, affecting biodiversity, including habitat for rare and endangered species, fire control, hiking, aesthetic enjoyment and tourism.

D. Adequacy of the Current Standards

An important issue to be addressed in the current review of the secondary standards for oxides of nitrogen and sulfur is whether, in view of the scientific evidence reflected in the ISA, additional information on exposure and risk discussed in the REA, and conclusions drawn from the PA, the existing standards provide adequate protection. The Administrator therefore, has

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considered the extent to which the current standards are adequate for the protection of public welfare. Having reached the general conclusion that aquatic and terrestrial ecosystems can be degraded by deposition of oxides of nitrogen and sulfur, it is then necessary to first evaluate the appropriateness (in terms of form and structure) of the current standards to address the ecological effects of oxides of nitrogen and sulfur as well as the adequacy of the current secondary standards for oxides of nitrogen and sulfur to provide requisite protection by considering to what degree risks to sensitive ecosystems would be expected to occur in areas that meet the current standards. Conclusions regarding the adequacy of the current standards are based on the available ecological effects, exposure and risk-based evidence. In evaluating the strength of this information, EPA has taken into account the uncertainties and limitations in the scientific evidence. This section addresses the adequacy of the current standards to protect against direct exposure effects on plants from oxides of nitrogen and sulfur, the appropriateness of the current structure of the standards to address deposition-related effects of oxides of nitrogen and sulfur on sensitive ecosystems and finally, the adequacy of such standards to protect against adverse effects related to the deposition of oxides of nitrogen and sulfur.

1. *Adequacy of the Current Standards for Direct Effects*

The current secondary oxides of nitrogen and sulfur standards are intended to protect against adverse effects to public welfare. For oxides of nitrogen, the current secondary standard was set identical to the primary standard[3], i.e., an annual standard set for NO_2 to protect against adverse effects on vegetation from direct exposure to ambient oxides of nitrogen. For oxides of sulfur, the current secondary standard is a 3-hour standard intended to provide protection for

[3] The current primary NO_2 standard has recently been changed to the 3-year average of the 98th percentile of the annual distribution of the 1 hour daily maximum of the concentration of NO_2. The current secondary standard remains as it was set in 1971.

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plants from the direct foliar damage associated with atmospheric concentrations of SO_2. It is appropriate to consider whether the current standards are adequate to protect against the direct effects on vegetation resulting from ambient NO_2 and SO_2 which were the basis for the current secondary standards. The ISA concluded that there was sufficient evidence to infer a causal relationship between exposure to SO_2, NO, NO_2 and PAN and injury to vegetation. Additional research on acute foliar injury has been limited and there is no evidence to suggest foliar injury below the levels of the current secondary standards for oxides of nitrogen and sulfur. There is sufficient evidence to suggest that the levels of the current standards are likely adequate to protect against direct phytotoxic effects.

2. *Appropriateness and Adequacy of the Current Standards for Deposition-related Effects*

This section addresses two concepts necessary to evaluate the current standards in the context of deposition related effects. First, appropriateness of the current standards is considered with regard to indicator, form, level and averaging time. This discussion centers around the ability of the current standards to evaluate and provide protection against deposition related effects that vary spatially and temporally. It includes particular emphasis on the indicators and forms of the current standards and the degree to which they are ecologically relevant with regard to deposition related effects. Second, this section evaluates the current standards in terms of adequacy of protection.

a. *Appropriateness*

The ISA has established that the major effects of concern for this review of the oxides of nitrogen and sulfur standards are associated with deposition of nitrogen and sulfur caused by atmospheric concentrations of oxides of nitrogen and sulfur. The current standards are not directed toward depositional effects, and none of the elements of the current NAAQS – indicator,

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form, averaging time, and level – are suited for addressing the effects of nitrogen and sulfur deposition.

Five issues arise that call into question the ecological relevance of the structure of the current secondary standards for oxides of nitrogen and sulfur.

(1) The current SO_2 secondary standard (0.5 ppm SO_2 over a 3-hour average) does not utilize an exposure period that is relevant for ecosystem impacts. The majority of deposition related impacts are associated with depositional loads that occur over periods of months to years. This differs significantly from exposures associated with hourly concentrations of SO_2 as measured by the current secondary standard. By addressing short-term concentrations, the current SO_2 secondary standard, while protective against direct foliar effects from gaseous oxides of sulfur, does not take into account the findings of effects in the ISA, which notes the relationship between annual deposition of sulfur and acidification effects which are likely to be more severe and widespread than phytotoxic effects under current ambient conditions, and include effects from long term deposition as well as short term. Acidification is a process that occurs over time because the ability of an aquatic system to counteract acidic inputs is reduced as natural buffers are used more rapidly than they can be replaced through geologic weathering. The relevant period of exposure for ecosystems is, therefore, not the exposures captured in the short averaging time of the current SO_2 secondary standard. The current secondary standard for oxides of nitrogen is an annual standard (0.053 ppm averaged over 1 year) and as such is more ecologically relevant.

(2) Current standards do not utilize appropriate atmospheric indicators. Nitrogen dioxide and SO_2 are used as the component of oxides of nitrogen and sulfur that are measured, but they do not provide a complete link to the direct effects on ecosystems from deposition of oxides

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of nitrogen and sulfur as they do not capture all relevant chemical species of oxidized nitrogen and oxidized sulfur that contribute to deposition. The ISA provides evidence that deposition related effects are linked with total nitrogen and total sulfur deposition, and thus all forms of oxidized nitrogen and oxidized sulfur that are deposited will contribute to effects on ecosystems. Thus, by using atmospheric NO_2 and SO_2 concentrations as indicators, the current standards address only a fraction of total atmospheric oxides of nitrogen and sulfur, and do not take into account the effects from deposition of total atmospheric oxides of nitrogen and sulfur. This suggests that more comprehensive atmospheric indicators should be considered in designing ecologically relevant standards.

(3) Current standards reflect separate assessments of the two individual pollutants, NO_2 and SO_2, rather than assessing the joint impacts of deposition to ecosystems. Recognizing the role that each pollutant plays in jointly affecting ecosystem indicators, functions, and services is vital to developing a meaningful standard. The clearest example of this interaction is in assessment of the impacts of acidifying deposition on aquatic ecosystems. Acidification in an aquatic ecosystem depends on the total acidifying potential of the deposition of both nitrogen and sulfur from both atmospheric deposition of oxides of nitrogen and sulfur as well as the inputs from other sources of nitrogen and sulfur such as reduced nitrogen and non-atmospheric sources. It is the joint impact of the two pollutants that determines the ultimate effect on organisms within the ecosystem, and critical ecosystem functions such as habitat provision and biodiversity. Standards that are set independently are less able to account for the contribution of the other pollutant. This suggests that interactions between oxides of nitrogen and oxides of sulfur should be a critical element of the conceptual framework for ecologically relevant standards. There are also important interactions between oxides of

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nitrogen and sulfur and reduced forms of nitrogen, which also contribute to acidification and nutrient enrichment. It is important that the structure of the standards address the role of reduced nitrogen in determining the ecological effects resulting from deposition of atmospheric oxides of nitrogen and sulfur. Consideration will also have to be given to total loadings as ecosystems respond to all sources of nitrogen and sulfur.

(4) Current standards do not take into account variability in ecosystem sensitivity. Ecosystems are not uniformly distributed either spatially or temporally in their sensitivity to oxides of nitrogen and sulfur. Therefore, failure to account for the major determinants of variability, including geological and soil characteristics related to the sensitivity to acidification or nutrient enrichment as well as atmospheric and landscape characteristics that govern rates of deposition, may lead to standards that do not provide requisite levels of protection across ecosystems. The current structures of the standards do not address the complexities in the responses of ecosystems to deposition of oxides of nitrogen and sulfur. Ecosystems contain complex groupings of organisms that respond in various ways to the alterations of soil and water that result from deposition of nitrogen and sulfur compounds. Different ecosystems therefore respond in different ways depending on a multitude of factors that control how deposition is integrated into the system. For example, the same levels of deposition falling on limestone dominated soils have a very different effect from those falling on shallow glaciated soils underlain with granite. One system may over time display no obvious detriment while the other may experience a catastrophic loss in fish communities. This degree of sensitivity is a function of many atmospheric factors that control rates of deposition as well as ecological factors that control how an ecosystem responds to that deposition. The current standards do not take into account spatial and seasonal variations, not only in

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depositional loadings, but also in sensitivity of ecosystems exposed to those loadings. Based on the discussion summarized above, the PA concludes that the current secondary standards for oxides of nitrogen and oxides of sulfur are not ecologically relevant in terms of averaging time, form, level or indicator.

b. *Adequacy of Protection*

As described in the PA, ambient conditions in 2005 indicate that the current SO_2 and NO_2 secondary standards were not exceeded at that time (US EPA, 2011, Figures 6-1 and 6-2) in locations where negative ecological effects have been observed. In many locations, SO_2 and NO_2 concentrations are substantially below the levels of the secondary standards. This pattern suggests that levels of deposition and any negative effects on ecosystems due to deposition of oxides of nitrogen and sulfur under recent conditions are occurring even though areas meet or are below current standards. In addition, based on conclusions in the REA, these levels will not decline in the future to levels below which it is reasonable to anticipate effects.

In determining the adequacy of the current secondary standards for oxides of nitrogen and sulfur the PA considered the extent to which ambient deposition contributes to loadings in ecosystems. Since the last review of the secondary standard for oxides of nitrogen, a great deal of information on the contribution of atmospheric deposition associated with ambient oxides of nitrogen has become available. The REA presents a thorough assessment of the contribution of oxidized nitrogen to nitrogen deposition throughout the U.S., and the relative contributions of ambient oxidized and reduced forms of nitrogen. The REA concludes that based on that analysis, ambient oxides of nitrogen are a significant component of atmospheric nitrogen deposition, even in areas with relatively high rates of deposition of reduced nitrogen. In

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addition, atmospheric deposition of oxidized nitrogen contributes significantly to total nitrogen loadings in nitrogen sensitive ecosystems.

The ISA summarizes the available studies of relative nitrogen contribution and finds that in much of the U.S., oxides of nitrogen contribute from 50 to 75 percent of total atmospheric deposition relative to total reactive nitrogen, which includes oxidized and reduced nitrogen species (US EPA, 2008, section 2.8.4). Although the proportion of total nitrogen loadings associated with atmospheric deposition of nitrogen varies across locations, the ISA indicates that atmospheric nitrogen deposition is the main source of new anthropogenic nitrogen to most headwater streams, high elevation lakes, and low-order streams. Atmospheric nitrogen deposition contributes to the total nitrogen load in terrestrial, wetland, freshwater and estuarine ecosystems that receive nitrogen through multiple pathways. In several large estuarine systems, including the Chesapeake Bay, atmospheric deposition accounts for between 10 and 40 percent of total nitrogen loadings (US EPA, 2008).

Atmospheric concentrations of oxides of sulfur account for nearly all sulfur deposition in the US. For the period 2004-2006, mean sulfur deposition in the U.S. was greatest east of the Mississippi River with the highest deposition amount, 21.3 kg S/ha-yr, in the Ohio River Valley where most recording stations reported 3-year averages >10 kg S/ha-yr. Numerous other stations in the East reported S deposition >5 kg S/ha-yr. Total sulfur deposition in the U.S. west of the 100th meridian was relatively low, with all recording stations reporting <2 kg S/ha-yr and many reporting <1 kg S/ha-yr. Sulfur was primarily deposited in the form of wet SO_4^{2-} followed in decreasing order by a smaller proportion of dry SO_2 and a much smaller proportion of deposition as dry SO_4^{2-}.

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As discussed throughout the REA (US EPA, 2009 and section II.B above), there are several key areas of risk that are associated with ambient concentrations of oxides of nitrogen and sulfur. As noted earlier, in previous reviews of the secondary standards for oxides of nitrogen and sulfur, the standards were designed to protect against direct exposure of plants to ambient concentrations of the pollutants. A significant shift in understanding of the effects of oxides of nitrogen and sulfur has occurred since the last reviews, reflecting the large amount of research that has been conducted on the effects of deposition of nitrogen and sulfur to ecosystems. The most significant current risks of adverse effects to public welfare are those related to deposition of oxides of nitrogen and sulfur to both terrestrial and aquatic ecosystems. These risks fall into two categories, acidification and nutrient enrichment, which were emphasized in the REA as most relevant to evaluating the adequacy of the existing standards in protecting public welfare from adverse ecological effects.

i. Aquatic acidification

The focus of the REA case studies was on determining whether deposition of sulfur and oxidized nitrogen in locations where ambient oxides of nitrogen and sulfur were at or below the current standards was resulting in acidification and related effects, including episodic acidification and mercury methylation. Based on the case studies conducted for lakes in the Adirondacks and streams in Shenandoah National Park (case studies are discussed more fully in section II.B and US EPA, 2009), there is significant risk to acid sensitive aquatic ecosystems at atmospheric concentrations of oxides of nitrogen and sulfur at or below the current standards. The REA also supports strongly a relationship between atmospheric deposition of oxides of nitrogen and sulfur and loss of ANC in sensitive ecosystems and indicates that ANC is an excellent indicator of aquatic acidification. The REA also concludes that at levels of deposition

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associated with oxides of nitrogen and sulfur concentrations at or below the current standards, ANC levels are expected to be below benchmark values that are associated with significant losses in fish species richness.

Significant portions of the U.S. are acid sensitive, and current deposition levels exceed those that would allow recovery of the most acid sensitive lakes in the Adirondacks (US EPA, 2008, Executive Summary). In addition, because of past loadings, areas of the Shenandoah are sensitive to current deposition levels (US EPA, 2008, Executive Summary). Parts of the West are naturally less sensitive to acidification and subjected to lower deposition (particularly SO_x) levels relative to the eastern United States, and as such, less focus in the ISA is placed on the adequacy of the existing standards in these areas, with the exception of the mountainous areas of the West, which experience episodic acidification due to deposition.

In describing the effects of acidification in the two case study areas the REA uses the approach of describing benchmarks in terms of ANC values. Many locations in sensitive areas of the U.S. have ANC levels below benchmark levels for ANC classified as severe, elevated, or moderate concern (US EPA, 2011, Figure 2-1). The average current ANC levels across 44 lakes in the Adirondack case study area is 62.1 μeq/L (moderate concern). However, 44 percent of lakes had deposition levels exceeding the critical load for an ANC of 50 μeq/L (elevated), and 28 percent of lakes had deposition levels exceeding the (higher) critical load for an ANC of 20 μeq/L (severe) (US EPA, 2009, section 4.2.4.2). This information indicates that almost half of the 44 lakes in the Adirondacks case study area are at an elevated concern level, and almost a third are at a severe concern level. These levels are associated with greatly diminished fish species diversity, and losses in the health and reproductive capacity of remaining populations. Based on assessments of the relationship between number of fish species and ANC level in both

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the Adirondacks and Shenandoah areas, the number of fish species is decreased by over half at an ANC level of 20 μeq/L relative to an ANC level at 100 μeq/L (US EPA, 2009, Figure 4.2-1). When extrapolated to the full population of lakes in the Adirondacks area using weights based on the EMAP probability survey (US EPA, 2009, section 4.2.6.1), 36 percent of lakes exceeded the critical load for an ANC of 50 μeq/L and 13 percent of lakes exceeded the critical load for an ANC of 20 μeq/L.

Many streams in the Shenandoah case study area also have levels of deposition that are associated with ANC levels classified as severe, elevated, or moderate concern. The average ANC under recent conditions for the 60 streams evaluated in the Shenandoah case study area is 57.9 μeq/L, indicating moderate concern. However, 85 percent of these streams had recent deposition exceeding the critical load for an ANC of 50 μeq/L, and 72 percent exceeded the critical load for an ANC of 20 μeq/L. As with the Adirondacks area, this information suggests that ANC levels may decline in the future and significant numbers of sensitive streams in the Shenandoah area are at risk of adverse impacts on fish populations if recent conditions persist. Many other streams in the Shenandoah area are also likely to experience conditions of elevated to severe concern based on the prevalence in the area of bedrock geology associated with increased sensitivity to acidification suggesting that effects due to stream acidification could be widespread in the Shenandoah area (US EPA, 2009, section 4.2.6.2).

In addition to these chronic acidification effects, the ISA notes that "consideration of episodic acidification greatly increases the extent and degree of estimated effects for acidifying deposition on surface waters" (US EPA, 2008, section 3.2.1.6). Some studies show that the number of lakes that could be classified as acid-impacted based on episodic acidification is 2 to 3 times the number of lakes classified as acid-impacted based on chronic ANC. These episodic

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acidification events can have long term effects on fish populations (US EPA, 2008, section 3.2.1.6). Under recent conditions, episodic acidification has been observed in locations in the eastern U.S. and in the mountainous western U.S. (US EPA, 2008, section 3.2.1.6).

The ISA, REA and PA all conclude that the current standards are not adequate to protect against the adverse impacts of aquatic acidification on sensitive ecosystems. A recent survey, as reported in the ISA, found sensitive streams in many locations in the U.S., including the Appalachian Mountains, the Coastal Plain, and the Mountainous West (US EPA, 2008, section 4.2.2.3). In these sensitive areas, between 1 and 6 percent of stream kilometers are chronically acidified. The REA further concludes that both the Adirondack and Shenandoah case study areas are currently receiving deposition from ambient oxides of nitrogen and sulfur in excess of their ability to neutralize such inputs. In addition, based on the current emission scenarios, forecast modeling out to the year 2020 as well as 2050 indicates a large number of streams in these areas will still be adversely impacted (section II.B). Based on these considerations, the PA concludes that the current secondary NAAQS for oxides of nitrogen and sulfur do not provide adequate protection of sensitive ecosystems with regard to aquatic acidification.

ii. Terrestrial acidification

Based on the terrestrial acidification case studies, Kane Experimental Forest in Pennsylvania and Hubbard Brook Experimental Forest described in section II.B) of sugar maple and red spruce habitat, the REA concludes that there is significant risk to sensitive terrestrial ecosystems from acidification at atmospheric concentrations of NO_2 and SO_2 at or below the current standards. The ecological indicator selected for terrestrial acidification is the BC/Al, which has been linked to tree health and growth. The results of the REA strongly support a relationship between atmospheric deposition of oxides of nitrogen and sulfur and BC/Al, and that

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BC/Al is a good indicator of terrestrial acidification. At levels of deposition associated with oxides of nitrogen and sulfur concentrations at or below the current standards, BC/Al levels are expected to be below benchmark values that are associated with significant effects on tree health and growth. Such degradation of terrestrial ecosystems could affect ecosystem services such as habitat provisioning, endangered species, goods production (timber, syrup, etc.) among others.

Many locations in sensitive areas of the U.S. have BC/Al levels below benchmark levels classified as providing low to intermediate levels of protection to tree health. At a BC/Al ratio of 1.2 (intermediate level of protection), red spruce growth can be reduced by 20 percent. At a BC/Al ratio of 0.6 (low level of protection), sugar maple growth can be decreased by 20 percent. The REA did not evaluate broad sensitive regions. However, in the sugar maple case study area (Kane Experimental Forest), recent deposition levels are associated with a BC/Al ratio below 1.2, indicating between intermediate and low level of protection, which would indicate the potential for a greater than 20 percent reduction in growth. In the red spruce case study area (Hubbard Brook Experimental Forest), recent deposition levels are associated with a BC/Al ratio slightly above 1.2, indicating slightly better than an intermediate level of protection (US EPA, 2009, section 4.3.5.1).

Over the full range of sugar maple, 12 percent of evaluated forest plots exceeded the critical loads for a BC/Al ratio of 1.2, and 3 percent exceeded the critical load for a BC/Al ratio of 0.6. However, there was large variability across states. In New Jersey, 67 percent of plots exceeded the critical load for a BC/Al ratio of 1.2, while in several states on the outskirts of the range for sugar maple (e.g. Arkansas, Illinois) no plots exceeded the critical load for a BC/Al ratio of 1.2. For red spruce, overall 5 percent of plots exceeded the critical load for a BC/Al ratio of 1.2, and 3 percent exceeded the critical load for a BC/Al ratio of 0.6. In the major red spruce

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producing states (Maine, New Hampshire, and Vermont), critical loads for a BC/Al ratio of 1.2 were exceeded in 0.5, 38, and 6 percent of plots, respectively.

The ISA, REA and PA all conclude that the current standards are not adequate to protect against the adverse impacts of terrestrial acidification on sensitive ecosystems. As stated in the REA and PA, the main drawback, with the understanding of terrestrial acidification lies in the sparseness of available data by which we can predict critical loads and that the data are based on laboratory responses rather than field measurements. Other stressors that are present in the field but that are not present in the laboratory may confound this relationship. The REA does however, conclude that the case study results, when extended to a 27 state region, show that nitrogen and sulfur acidifying deposition in the sugar maple and red spruce forest areas caused the calculated Bc/Al ratio to fall below 1.2 (the intermediate level of protection) in 12 percemt of the sugar maple plots and 5 percent of the red spruce plots; however, results from individual states ranged from 0 to 67 percent of the plots for sugar maple and 0 to 100 percent of the plots for red spruce.

iii. Terrestrial nutrient enrichment

Nutrient enrichment effects are due to nitrogen loadings from both atmospheric and non-atmospheric sources. Evaluation of nutrient enrichment effects requires an understanding that nutrient inputs are essential to ecosystem health and that specific long term levels of nutrients in a system affect the types of species that occur over long periods of time. Short term additions of nutrients can affect species competition, and even small additions of nitrogen in areas that are traditionally nutrient poor can have significant impacts on productivity as well as species composition. Most ecosystems in the U.S. are nitrogen-limited, so regional decreases in emissions and deposition of airborne nitrogen compounds could lead to some decrease in growth

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of the vegetation that surrounds the targeted aquatic system but as discussed below evidence for this is mixed. Whether these changes in plant growth are seen as beneficial or adverse will depend on the nature of the ecosystem being assessed.

Information on the effects of changes in nitrogen deposition on forestlands and other terrestrial ecosystems is very limited. The multiplicity of factors affecting forests, including other potential stressors such as ozone, and limiting factors such as moisture and other nutrients, confound assessments of marginal changes in any one stressor or nutrient in forest ecosystems. The ISA notes that only a fraction of the deposited nitrogen is taken up by the forests, most of the nitrogen is retained in the soils (US EPA, 2008, section 3.3.2.1). In addition, the ISA indicates that forest management practices can significantly affect the nitrogen cycling within a forest ecosystem, and as such, the response of managed forests to nitrogen deposition will be variable depending on the forest management practices employed in a given forest ecosystem (US EPA, 2008, Annex C C.6.3). Increases in the availability of nitrogen in nitrogen-limited forests via atmospheric deposition could increase forest production over large non-managed areas, but the evidence is mixed, with some studies showing increased production and other showing little effect on wood production (US EPA, 2008, section 3.3.9). Because leaching of nitrate can promote cation losses, which in some cases create nutrient imbalances, slower growth and lessened disease and freezing tolerances for forest trees, the net effect of increased N on forests in the U.S. is uncertain (US EPA, 2008, section 3.3.9).

The scientific literature has many examples of the deleterious effects caused by excessive nitrogen loadings to terrestrial systems. Several studies have set benchmark values for levels of N deposition at which scientifically adverse effects are known to occur. Large areas of the country appear to be experiencing deposition above these benchmarks. The ISA indicates

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studies that have found that at 3.1 kg N/ha/yr, the community of lichens begins to change from acidophytic to tolerant species; at 5.2 kg N/ha/yr, the typical dominance by acidophytic species no longer occurs; and at 10.2 kg N/ha/yr, acidophytic lichens are totally lost from the community. Additional studies in the Colorado Front Range of the Rocky Mountain National Park support these findings. These three values (3.1, 5.2, and 10.2 kg/ha/yr) are one set of ecologically meaningful benchmarks for the mixed conifer forest (MCF) of the pacific coast regions. Nearly all of the known sensitive communities receive total nitrogen deposition levels above the 3.1 N kg/ha/yr ecological benchmark according to the12 km, 2002 CMAQ/NADP data, with the exception of the easternmost Sierra Nevadas. The MCFs in the southern portion of the Sierra Nevada forests and nearly all MCF communities in the San Bernardino forests receive total nitrogen deposition levels above the 5.2 N kg/ha/yr ecological benchmark.

Coastal Sage Scrub communities are also known to be sensitive to community shifts caused by excess nitrogen loadings. Studies have investigated the amount of nitrogen utilized by healthy and degraded CSS systems. In healthy stands, the authors estimated that 3.3 kg N/ha/yr was used for CSS plant growth. It is assumed that 3.3 kg N/ha/yr is near the point where nitrogen is no longer limiting in the CSS community and above which level community changes occur, including dominance by invasive species and loss of coastal sage scrub. Therefore, this amount can be considered an ecological benchmark for the CSS community. The majority of the known CSS range is currently receiving deposition in excess of this benchmark. Thus, the REA concludes that recent conditions where oxides of nitrogen ambient concentrations are at or below the current oxides of nitrogen secondary standards are not adequate to protect against anticipated adverse impacts from N nutrient enrichment in sensitive ecosystems.

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iv. Aquatic nutrient enrichment

The REA aquatic nutrient enrichment case studies focused on coastal estuaries and revealed that while current ambient loadings of atmospheric oxides of nitrogen are contributing to the overall depositional loading of coastal estuaries, other non-atmospheric sources are contributing in far greater amounts in total, although atmospheric contributions are as large as some other individual source types. The ability of current data and models to characterize the incremental adverse impacts of nitrogen deposition is limited, both by the available ecological indicators, and by the inability to attribute specific effects to atmospheric sources of nitrogen. The REA case studies used ASSETS EI as the ecological indicator for aquatic nutrient enrichment. This index is a six level index characterizing overall eutrophication risk in a water body. This indictor is not sensitive to changes in nitrogen deposition within a single level of the index. In addition, this type of indicator does not reflect the impact of nitrogen deposition in conjunction with other sources of nitrogen.

Based on the above considerations, the REA concludes that the ASSETS EI is not an appropriate ecological indicator for estuarine aquatic eutrophication and that additional analysis is required to develop an appropriate indicator for determining the appropriate levels of protection from N nutrient enrichment effects in estuaries related to deposition of oxides of nitrogen. As a result, EPA is unable to make a determination as to the adequacy of the existing secondary oxides of nitrogen standard in protecting public welfare from nitrogen nutrient enrichment effects in estuarine aquatic ecosystems.

Additionally, nitrogen deposition can alter species composition and cause eutrophication in freshwater systems. In the Rocky Mountains, for example, deposition loads of 1.5 to 2 kg/ha/yr which are well within current ambient levels are known to cause changes in species

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composition in diatom communities indicating impaired water quality (US EPA, 2008, section 3.3.5.3). This suggests that the existing secondary standard for oxides of nitrogen does not protect such ecosystems and their resulting services from impairment.

v. Other effects

An important consideration in looking at the effects of deposition of oxides of sulfur in aquatic ecosystems is the potential for production of MeHg, a neurotoxic contaminant. The production of meaningful amounts of MeHg requires the presence of SO_4^{2-} and mercury, and where mercury is present, increased availability of SO_4^{2-} results in increased production of MeHg. There is increasing evidence on the relationship between sulfur deposition and increased methylation of mercury in aquatic environments; this effect occurs only where other factors are present at levels within a range to allow methylation. The production of MeHg requires the presence of SO_4^{2-} and mercury, but the amount of MeHg produced varies with oxygen content, temperature, pH and supply of labile organic carbon (US EPA, 2008, section 3.4). In watersheds where changes in sulfate deposition did not produce an effect, one or several of those interacting factors were not in the range required for meaningful methylation to occur (US EPA, 2008, section 3.4). Watersheds with conditions known to be conducive to mercury methylation can be found in the northeastern United States and southeastern Canada (US EPA, 2009, section 6).

With respect to sulfur deposition and mercury methylation, the final ISA determined that ”[t]he evidence is sufficient to infer a causal relationship between sulfur deposition and increased mercury methylation in wetlands and aquatic environments.” However, EPA did not conduct a quantitative assessment of the risks associated with increased mercury methylation under current conditions. As such, EPA is unable to make a determination as to the adequacy of the existing

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SO_2 secondary standards in protecting against welfare effects associated with increased mercury methylation.

vi. Summary of Adequacy Considerations

In summary, the PA concludes that currently available scientific evidence and assessments clearly call into question the adequacy of the current standards with regard to deposition-related effects on sensitive aquatic and terrestrial ecosystems, including acidification and nutrient enrichment. Further, the PA recognizes that the elements of the current standards--indicator, averaging time, level and form – are not ecologically relevant, and are thus not appropriate for standards designed to provide such protection. Thus, the PA concludes that consideration should be given to establishing a new ecologically relevant multi-pollutant, multimedia standard to provide appropriate protection from deposition-related ecological effects of oxides of nitrogen and sulfur on sensitive ecosystems, with a focus on protecting against adverse effects associated with acidifying deposition in sensitive aquatic ecosystems.

3. *CASAC Views*

In a letter to the Administrator (Russell and Samet 2011a), the CASAC Oxides of Nitrogen and Oxides of Sulfur Panel, with full endorsement of the chartered CASAC, unanimously concluded that:

> EPA staff has demonstrated through the Integrated Science Assessment (ISA), Risk and Exposure Characterization (REA) and the draft PA that ambient NOx and SOx can have, and are having, adverse environmental impacts. The Panel views that the current NOx and SOx secondary standards should be retained to protect against direct adverse impacts to vegetation from exposure to gas phase exposures of these two families of air pollutants. Further, the ISA, REA and draft PA demonstrate that adverse impacts to aquatic ecosystems are also occurring due to deposition of NOx and SOx. Those impacts include acidification and undesirable levels of nutrient enrichment in some aquatic ecosystems. The levels of the current NOx and SOx secondary NAAQS are not sufficient, nor the forms

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of those standards appropriate, to protect against adverse depositional effects; thus a revised NAAQS is warranted.

In addition, with regard to the joint consideration of both oxides of nitrogen and oxides of sulfur as well as the consideration of deposition related effects, CASAC concluded that the PA had developed a credible methodology for considering such effects. The Panel stated that "the Policy Assessment develops a framework for a multi-pollutant, multimedia standard that is ecologically relevant and reflects the combined impacts of these two pollutants as they deposit to sensitive aquatic ecosystems."

4. *Administrator's Proposed Conclusions Concerning Adequacy of Current Standard*

Based on the above considerations and taking into account CASAC advice, the Administrator recognizes that the purpose of the secondary standard is to protect against "adverse" effects resulting from exposure to oxides of nitrogen and sulfur, discussed above in section II.A. The Administrator also recognizes the need for conclusions as to the adequacy of the current standards for both direct and deposition related effects as well as conclusions as to the appropriateness and ecological relevance of the current standards.

In considering what constitutes an ecological effect that is also adverse to the public welfare, the Administrator took into account the ISA conclusions regarding the nature and strength of the effects evidence, the risk and exposure assessment results, the degree to which the associated uncertainties should be considered in interpreting the results, the conclusions presented in the PA, and the views of CASAC and members of the public. On these bases, the Administrator concludes that the current secondary standards are adequate to protect against direct phytotoxic effects on vegetation. Thus, the Administrator proposes to retain the current

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secondary standard for oxides of nitrogen at 53 ppb,[4] annual average concentration, measured in the ambient air as NO_2, and the current secondary standard for oxides of sulfur at 0.5 ppm, 3-hour average concentration, measured in the ambient air as SO_2.

With regard to deposition-related effects, the Administrator has first to consider the appropriateness of the structure of the current standards to address ecological effects of concern. Based on the evidence as well as considering the advice given by CASAC on this matter, the Administrator concludes that the elements of the current standards are not ecologically relevant and thus are not appropriate to provide protection of ecosystems. On the subject of adequacy of protection with regard to deposition-related effects, the Administrator considered the full nature of ecological effects related to the deposition of ambient oxides of nitrogen and sulfur into sensitive ecosystems across the country. Her conclusions are based on the evidence presented in the ISA with regard to acidification and nutrient enrichment effects, the findings of the REA with regard to scope and severity of the current and likely future effects of deposition, the synthesis of both the scientific evidence and risk and exposure results in the PA as to the adequacy of the current standards, and the advice of both CASAC and the public. After such consideration, the Administrator concludes that current levels of oxides of nitrogen and sulfur are sufficient to cause acidification of both aquatic and terrestrial ecosystems, nutrient enrichment of terrestrial ecosystems and contribute to nutrient enrichment effects in estuaries that could be considered adverse, and the current secondary standards do not provide adequate protection from such effects.

[4] The annual secondary standard for oxides of nitrogen is being specified in units of ppb to conform to the current version of the annual primary standard, as specified in the final rule for the most recent review of the NO_2 primary NAAQS (75 FR 6531; February 9, 2010).

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Having reached these conclusions, the Administrator determines that it is appropriate to consider alternative standards that are ecologically relevant. These considerations support the conclusion that the current secondary standards is neither appropriate nor adequate to protect against deposition related effects. The Administrator's consideration of such alternative standards is discussed below in Section III.

III. Rationale for Proposed Decision on Alternative Multi-pollutant Approach to Secondary Standards for Aquatic Acidification

Having reached the conclusion that the current NO_2 and SO_2 secondary standards are not adequate to provide appropriate protection against deposition-related effects associated with oxides of nitrogen and sulfur, the Administrator then considered what new multi-pollutant standard might be appropriate, at this time, to address such effects on public welfare. The Administrator recognizes that the inherently complex and variable linkages between ambient concentrations of nitrogen and sulfur oxides, the related deposited forms of nitrogen and sulfur, and the ecological responses that are associated with public welfare effects call for consideration of an ecologically relevant design of a standard that reflects these linkages. The Administrator also recognizes that characterization of such complex and variable linkages will necessarily require consideration of information and analyses that have important limitations and uncertainties.

Despite its complexity, an ecologically relevant multi-pollutant standard to address deposition-related effects could still appropriately be defined in terms of the same basic elements that are used to define any NAAQS – indicator, form, averaging time, and level. The form would incorporate additional structural elements that reflect relevant multi-pollutant and multimedia attributes. These structural elements include the use of an ecological indicator, tied

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to the ecological effect we are focused on, and other elements that account for ecologically relevant factors other than ambient air concentrations. All of these elements would be needed to enable a linkage from ambient air indicators to the ecological indicator to define an ecologically relevant standard. As a result, such a standard would necessarily be more complex than the NAAQS that have been set historically to address effects associated with ambient concentrations of a single pollutant.

More specifically, the Administrator considered an ecologically relevant multi-pollutant standard to address effects associated with acidifying deposition related to ambient concentrations of oxides of nitrogen and sulfur in sensitive aquatic ecosystems. This focus is consistent with the information presented in the ISA, REA, and PA, which highlighted the sufficiency of the quantity and quality of the available evidence and assessments associated with aquatic acidification relative to the information and assessments available for other deposition-related effects, including terrestrial acidification and aquatic and terrestrial nutrient enrichment. Based on its review of these documents, CASAC agreed that aquatic acidification should be the focus for developing a new multi-pollutant standard in this review. In reaching conclusions about an air quality standard designed to address deposition-related aquatic acidification effects, the Administrator also recognizes that such a standard may also provide some degree of protection against other deposition-related effects.

As discussed in chapter 7 of the PA, the development of a new multi-pollutant standard to address deposition-related aquatic acidification effects recognizes the need for consideration of a nationally applicable standard for protection against adverse effects of aquatic acidification on public welfare, while recognizing the complex and heterogeneous interactions between ambient air concentrations of nitrogen and sulfur oxides, the related deposition of nitrogen and sulfur, and

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associated ecological responses. The development of such a standard also needs to take into account the limitations and uncertainties in the available information and analyses upon which characterization of such interactions are based. The approach used in the PA also recognizes that while such a standard would be national in scope and coverage, the effects to public welfare from aquatic acidification will not occur to the same extent in all locations in the U.S., given the inherent variability of the responses of aquatic systems to the effects of acidifying deposition.

As discussed above in section II, many locations in the U.S. are naturally protected against acid deposition due to underlying geological conditions. Likewise, some locations in the U.S., including lands managed for commercial agriculture and forestry, are not likely to be negatively impacted by current levels of nitrogen and sulfur deposition. As a result, while a new ecologically relevant secondary standard would apply everywhere, it would be structured to account for differences in the sensitivity of ecosystems across the country. This would allow for appropriate protection of sensitive aquatic ecosystems, which are relatively pristine and wild and generally in rural areas, and the services provided by such sensitive ecosystems, without requiring more protection than is needed elsewhere.

As discussed below, the multi-pollutant standard developed in the PA would employ (1) total reactive oxidized nitrogen (NO_y) and SO_x as the atmospheric ambient air indicators; (2) a form that takes into account variable factors, such as atmospheric and ecosystem conditions that modify the amounts of deposited nitrogen and sulfur; the distinction between oxidized and reduced forms of nitrogen; effects of deposited nitrogen and sulfur on aquatic ecosystems in terms of the ecological indicator ANC; and the representativeness of water bodies within a defined spatial area; (3) a multi-year averaging time, and (4) a standard level defined in terms of a single, national target ANC value that, in the context of the above form, identifies the levels of

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concentrations of NO_y and SO_x in the ambient air that would meet the standard. The form of such a standard has been defined by an index, AAI, which reflects the relationship between ambient concentrations of NO_y and SO_x and aquatic acidification effects that result from nitrogen and sulfur deposition related to these ambient concentrations.

In presenting the considerations associated with such an air quality standard to address deposition-related aquatic acidification effects, the following sections focus on each element of the standard, including indicator (section III.A), form (section III.B), averaging time (section III.C), and level (section III.D). Alternative combinations of levels and forms are discussed in section III.E. Considerations related to important uncertainties inherent in such an approach are discussed in section III.F. Advice from CASAC on such a new standard is presented in section III.G. The Administrator's proposed decisions on such a new standard are presented in section III.H.

A. Ambient Air Indicators

In considering alternative ambient air indicators, the PA primarily focuses on the important attribute of association. Association in a broad sense refers to how well an ambient air indicator relates to the ecological effects of interest by virtue of both the framework that links the ambient indicator and effects and the empirical evidence that quantifies the linkages. The PA also considers how measurable or quantifiable an indicator is to enable its use as an effective indicator of relevant ambient air concentrations.

As discussed above in section II.C, the PA concludes that indicators other than NO_2 and SO_2 should be considered as the appropriate indicators of oxides of nitrogen and sulfur in the ambient air for protection against the acidification effects associated with deposition of the associated nitrogen and sulfur. This conclusion is based on the recognition that all forms of

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nitrogen and sulfur in the ambient air contribute to deposition and resulting acidification, and as such, NO_2 and SO_2 are incomplete indicators. In principle, the ambient indicators should represent the species that are associated with oxides of nitrogen and sulfur in the ambient air and can contribute acidifying deposition. This includes both the species of oxides of nitrogen and sulfur that are directly emitted as well as species transformed in the atmosphere from oxides of nitrogen and sulfur that retain the nitrogen and sulfur atoms from directly emitted oxides of nitrogen and sulfur. All of these compounds are associated with oxides of nitrogen and sulfur in the ambient air and can contribute to acidifying deposition.

The PA focuses in particular on the various compounds with nitrogen or sulfur atoms that are associated with oxides of nitrogen and sulfur, because the acidifying potential is specific to nitrogen and sulfur, and not other atoms (e.g., H, C, O) whether derived from the original source of oxides of nitrogen and sulfur emissions or from atmospheric transformations. For example, the acidifying potential of each molecule of NO_2, NO, HNO_3 or PAN is identical, as is the potential for each molecule of SO_2 or ion of particulate sulfate, p-SO_4. Each atom of sulfur affords twice the acidifying potential of each atom of nitrogen.

1. Oxides of Sulfur

As discussed in the PA (US EPA, 2011, section 7.1.1), oxides of sulfur include the gases sulfur monoxide (SO), SO_2, sulfur trioxide (SO_3), disulfur monoxide (S_2O), and particulate-phase sulfur compounds (referred to as SO_4) that result from gas-phase sulfur oxides interacting with particles. However, the sum of SO_2 and SO_4 does represent virtually the entire ambient air mass of sulfur that contributes to acidification. In addition to accounting for virtually all the potential for acidification from oxidized sulfur in the ambient air, there are reliable methods to monitor the concentrations of SO_2 and particulate SO_4. In addition, much of the data used to

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develop the technical basis for the standard developed in the PA is based on monitoring or modeling of these species.[5] The PA concludes that the sum of SO_2 and SO_4, referred to as SO_x, are appropriate ambient air indicators of oxides of sulfur because they represent virtually all of the acidification potential of ambient air oxides of sulfur and there are reliable methods suitable for measuring SO_2 and SO_4.

2. Oxides of Nitrogen

As discussed in the PA (US EPA, 2011,section 7.1.2), NO_y, as defined in chapter 2 of the PA, incorporates basically all of the oxidized nitrogen species that have acidifying potential and as such, NO_y should be considered as an appropriate indicator for oxides of nitrogen. Total reactive oxidized nitrogen is an aggregate measure of NO and NO_2 and all of the reactive oxidized products of NO and NO_2. That is, NO_y is a group of nitrogen compounds in which all of the compounds are either an oxide of nitrogen or compounds in which the nitrogen atoms came from oxides of nitrogen. Total reactive oxidized nitrogen is especially relevant as an ambient indicator for acidification in that it both relates to the oxides of nitrogen in the ambient air and also represents the acidification potential of all oxidized nitrogen species in the ambient air, whether an oxide of nitrogen or derived from oxides of nitrogen.

There are currently available reliable methods of measuring aggregate NO_y. The term "aggregate" measure means that the NO_y, as measured, is not based on measuring each individual species of NO_y and calculating an NO_y value by summing the individual species. Rather, as described in chapter 2 of the PA, current measurement techniques process all of the individual NO_y species to produce a single aggregate measure of all of the nitrogen atoms

[5] As discussed in chapter 2 of the PA, SO_2 and particulate SO_4 are routinely measured in ambient air monitoring networks, although only the Clean Air Status and Trends Network (CASTNET) filter packs do not intentionally exclude particle size fractions. The CMAQ treatment of SOx is the simple addition of both species, which are treated explicitly in the model formulation. All particle size fractions are included in the CMAQ SOx estimates.

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associated with any NO_y species. Consequently, the NO_y measurement effectively provides the sum of all individual species, but the identity of the individual species is lost. As discussed above, the accounting for the individual nitrogen atoms is an accounting of the ambient air acidification potential of oxides of nitrogen and their transformation products and therefore the most relevant ambient indicator for aquatic acidification effects associated with oxides of nitrogen.

This loss of the information on individual species motivated consideration of alternative or more narrowly defined indicators for oxides of nitrogen in the PA. Consideration of a subset of NO_y species was based on the following reasoning. First, the actual dry deposition of nitrogen is determined on an individual species basis by multiplying the species concentration times a species-specific deposition velocity and then summed to develop an estimate of total dry deposition. Consequently, the use of individual ambient species has the potential to be more consistent with the underlying science of deposition and, therefore, has the potential to allow for a more rigorous evaluation of dry deposition with specialized field studies. In addition, there has been a suggestion of focusing only on the most quickly depositing NO_yspecies, such as HNO_3, as contributions from other NO_y species such as NO_2 may be negligible. These alternative indicators are discussed below.

The PA considers the relative merits of using each individual NO_y species as part of a group of indicators. In so doing, it was first noted that dry deposition of NO_y is treated as the sum of the deposition of each individual species in advanced process-based air quality models like CMAQ, as described in chapter 2 of the PA. Conceptually one could extend this process-based approach by using all NO_y species individually as separate indicators for oxides of nitrogen and requiring, for example, measurements of each of the species, including the

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dominant species of HNO_3, particulate nitrate (p-NO_3), true NO_2, NO and PAN. The potential attraction of using individual species would be the reliance on actual deposition velocities. This could have more physical meaning in comparison to a constructed model of aggregate deposition of NO_y, which is difficult to evaluate with observations because of the assimilation of many species with disparate deposition behavior. The PA notes that the major drawback of using individual NO_y species as the indicators is the lack of reliable measurement techniques, especially for PAN and NO_2 in rural locations, which renders the use of virtually any individual NO_y species, except for NO and perhaps p-NO_3, as functionally inadequate from a measurement perspective.

The PA next considered the relative merits of using a subset of NO_y species as the indicators for oxides of nitrogen, as was discussed above for oxides of sulfur. To the extent that certain species provide relatively minor contributions to total NO_y deposition, it may be appropriate to consider excluding them as part of the indicator. As discussed in chapter 2 of the PA, each nitrogen species within the array of NO_y species has species-specific dry deposition velocities. For example, the deposition velocity of HNO_3 is much greater than the velocity for NO_2 and, consequently, for a similar ambient air concentration, HNO_3 contributes more deposition of acidifying nitrogen relative to NO_2. In transitioning from source-oriented urban locations to rural environments, the ratio of the concentrations of HNO_3 and PAN to NO_2 increases.

Based on the reasoning that a larger fraction of the deposited NO_y is accounted for by total nitrate (the sum of HNO_3 and p-NO_3), a surrogate for the more rapidly depositing fraction of NO_y, combined with the availability of reliable total nitrate measurements through the CASTNET, the PA considered using total nitrate as the indicator for oxides of nitrogen (US

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EPA, 2011, appendix E). Nitrate would be expected to correlate well with total reactive oxidized nitrogen deposition relative to NO_y (US EPA, 2011, chapter 2) despite the inherent noise associated with variable contributions of low deposition velocity species (e.g., NO_2) that may have relatively high ambient concentrations. However, modeling simulations suggest that NO_y may be a more robust indicator, relative to HNO_3, in terms of relating absolute changes in ambient air concentrations to changes in nitrogen deposition driven by changes in ambient concentrations of oxides of nitrogen (US EPA, 2011, Figure 2-32).

Based on the above considerations, the PA concludes that NO_y should be considered as the appropriate ambient indicator for oxides of nitrogen based on its direct relationship to oxides of nitrogen in the ambient air and its direct relationship to deposition associated with aquatic acidification. Because NO_y represents all of the potentially acidifying oxidized nitrogen species in the ambient air, it is appropriately associated with the deposition of potentially acidifying compounds associated with oxides of nitrogen in the ambient air. In addition, there are reliable methods available to measure NO_y. Measurement of each individual species of NO_y, or the measurement of only a subset of species of NO_y, is less appropriate because there are not reliable measurements methods available to measure all of the individual species of NO_y and a subset of species would fail to account for significant portions of the oxidized reactive nitrogen that relate to acidification.[6]

B. Form

Based on the evidence of the aquatic acidification effects caused by the deposition of NO_y and SO_x, the PA (US EPA, 2011,section 7.2) presents the development of a new form that is

[6] The PA also notes that NO_y is a useful measurement for model evaluation purposes, which is especially important, recognizing the unique role that CMAQ plays in the development of this standard, as described below in section III.B.

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ecologically relevant for addressing such effects. The conceptual design for the form of such a standard includes three main components: an ecological indicator, deposition metrics that relate to the ecological indicator, and a function that relates ambient air indicators to deposition metrics. Collectively, these three components link the ecological indicator to ambient air indicators, as illustrated above in Fig II-1.

The simplified flow diagram in Figure II-1 compresses the various atmospheric, biological, and geochemical processes associated with acidifying deposition to aquatic ecosystems into a simplified conceptual picture. The ecological indicator (left box) is related to atmospheric deposition through biogeochemical ecosystem models (middle box), which associate a target deposition load to a target ecological indicator. Once a target deposition is established, associated allowable air concentrations are determined (right box) through the relationships between concentration and deposition that are embodied in air quality models such as CMAQ. The following discussion describes the development and rationale for each of these components, as well as the integration of these components into the full expression of the form of the standard using the concept of a national AAI that represents a target ANC level as a function of ambient air concentrations. Spatial aggregation issues associated with defining each of the terms of this index are also addressed below.

The AAI is designed to be an ecologically relevant form of the standard that determines the levels of NOy and SOx in the ambient air that would achieve a target ANC limit for the U.S. The intent of the AAI is to weight atmospheric concentrations of oxides of nitrogen and sulfur by their propensity to contribute to acidification through deposition, given the fundamental acidifying potential of each pollutant, and to take into account the ecological factors that govern acid sensitivity in different ecosystems. The index also accounts for the contribution of reduced

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nitrogen to acidification. Thus, the AAI encompasses those attributes of specific relevance to protecting ecosystems from the acidifying potential of ambient air concentrations of NOy and SOx.

1. Ecological Indicator

In considering alternative ecological indicators, the PA again primarily focuses on the attribute of association. In the case of an ecological indicator for aquatic acidification, association refers to the relationship between the indicator and adverse effects as discussed in section II. Because of the conceptual structure of the form of an AAI-based standard (Figure III-1), this particular ecological indicator must also link up in a meaningful and quantifiable manner with acidifying atmospheric deposition. In effect, the ecological indicator for aquatic acidification is the bridge between biological impairment and deposition of $NO_y$and SO_x.

This section presents the rationale in the PA for selecting ANC as the appropriate ecological indicator for consideration. Recognizing that ANC is not itself the causative or toxic agent for adverse aquatic acidification effects, the rationale for using ANC as the relevant ecological indicator is based on the following:

(1) The ANC is directly associated with the causative agents, pH and dissolved Al, both through empirical evidence and mechanistic relationships;

(2) Empirical evidence shows very clear and strong relationships between adverse effects and ANC;

(3) The ANC is a more reliable indicator from a modeling perspective, allowing use of a body of studies and technical analyses related to ANC and acidification to inform the development of the standard; and

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(4) The ANC literally embodies the concept of acidification as posed by the basic principles of acid base chemistry and the measurement method used to estimate ANC and, therefore, serves as a direct index to protect against acidification.

Ecological indicators of acidification in aquatic ecosystems can be chemical or biological components of the ecosystem that are altered by the acidifying effects of nitrogen and sulfur deposition. A desirable ecological indicator for aquatic acidification is one that is measurable or estimable, linked causally to deposition of nitrogen and sulfur, and linked causally, either directly or indirectly to ecological effects known or anticipated to adversely affect public welfare.

As summarized in chapter 2 of the PA, atmospheric deposition of NOy and SOx causes aquatic acidification through the input of strong acid anions (e.g., NO_3^- and SO_4^{2-}) that ultimately shifts the water chemistry equilibrium toward increased hydrogen ion levels (or decreased pH). The anions are deposited either directly to the aquatic ecosystem or indirectly via transformation through soil nitrification processes and subsequent drainage from terrestrial ecosystems. In other words, when these anions are mobilized in the terrestrial soil, they can leach into adjacent water bodies. Aquatic acidification is indicated by changes in the surface water chemistry of ecosystems. In turn, the alteration of surface water chemistry has been linked to negative effects on the biotic integrity of freshwater ecosystems. There is a suite of chemical indicators that could be used to assess the effects of acidifying deposition on lake or stream acid-base chemistry. These indicators include ANC; alkalinity (ALK); base neutralizing capacity, commonly referred to as acidity (ACY); surface water pH; concentrations of trivalent aluminum, Al^{+3}; and concentrations of major anions (SO_4^{2-}, NO_3^-), cations (Ca^{2+}, Mg^{+2}, K^+), or sums of cations or anions.

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The ANC and ALK are very similar quantities and are used interchangeably in the literature and for some of the analyses presented in this document. Both ANC and ALK are defined as the amount of strong acid required to reach a specified equivalence point. For acid-base solutions, an equivalence point can be thought of as the point at which the addition of strong acids (i.e., titration) is no longer neutralized by the solution. This explains the term acid neutralizing capacity, or ANC, as ANC relates directly to the capacity of a system to neutralize acids. The differences between ANC and ALK are based on operational definitions and subject to various interpretations. The ANC is preferred over ALK as the body of scientific evidence has focused on ANC and effects relationships. The ALK is more widely associated with more general characterizations of water quality such as the relative hardness of water associated with carbonates.

Indictors such as the concentrations of specific anions, cations, or their groupings, while relevant to acidification processes, are not robust acidification indicators as it is the relative balance of cations and anions that is more directly associated with acidification. That balance is captured by ANC and ALK. Acidity, ACY, is the converse of ANC and indicates how much strong base it takes to reach an equivalence point. Because ACY is not used in most ecosystem assessments, the body of information relating ACY to effects is too limited to serve as a basis for an appropriate ecological indicator. Aluminum and other metals are causative toxic agents that directly impair biological functions. However, Al, or metals in general, have high variability in concentrations that can be linked to effects, often at extremely low levels which in some cases approach detectability limits, exhibit rapid transient responses, and are often confounded by the presence of other toxic metals. These concerns limit the use of metals as reliable and measurable ecological indicators. Hydrogen ion (H^+) concentrations, using their negative logarithmic values,

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or pH, are well correlated with adverse effects, as discussed above in section II.A, and determine the solubility of metals such as aluminum. However, pH is not a preferred acidification indicator due to its highly transient nature and other concerns, as discussed below.

Having reasoned that ANC is a preferred indicator to ALK, ACY, individual metals or groupings of ions, the PA considers the relative merits of ANC compared to pH, which is a well recognized indicator of acidity and a more direct causative agent with regard to adverse effects. First, the linkage between ANC and pH is considered in recognition of the causative association between pH and effects.

The ANC is not the direct causative toxic agent impacting aquatic species diversity. The scientific literature generally emphasizes the links between pH and adverse effects as described above in section II.A. It is important, therefore, to consider the extent to which ANC and pH are well related from a mechanistic perspective as well as through empirical evidence. The ANC and pH are co-dependent on each other based on the requirement that all solutions are electrically neutral, meaning that any solution must satisfy the condition that all negatively charged species must be balanced by all positively charged species. The ANC is defined as the difference between strong anions and cations (US EPA, 2011, equation 7-13).

While the chemistry can be complex, the co-dependency between ANC and pH is explained by recognizing that positively charged hydrogen, H^+, is incorporated in the charge balance relationships related to the overall solution chemistry which also defines ANC. The positive, directional co-dependency (i.e., ANC and pH increase together) is further explained in concept as ANC reflects how much strong acid (i.e., how much hydrogen ion) it takes to titrate to an equivalence point. Strong observed correlations between pH and ANC as described in the PA support these mechanistic relationships.

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As discussed above in section II.A, there are well established examples of ANC correlating strongly with a variety of ecological effects which are summarized in the PA (US EPA, 2011, Table 3-1). Because pH and ANC are well correlated and linearly dependent over the pH ranges (4.5-6) where adverse ecological effects are observed, evidence of clear associations exist between ANC and adverse ecological effects as described in the PA. In large measure, this dependence between pH and ANC and the relationship of both pH and ANC to effects, speak directly to the appropriateness of ANC with respect to its use as an ecological indicator.

Thus, there is a clear association between ANC and ecological effects, although there is a more direct causal relationship between pH and ecological effects. Nonetheless, ANC is preferred as an ecological indicator based on its superior ability to provide a linkage with deposition in a meaningful and quantifiable manner, a role that is served far more effectively by ANC than by pH. While both ANC and pH are clearly associated with the effects of concern, ANC is superior in linking these effects to deposition.

The PA notes that the basis for this conclusion is that acidifying atmospheric deposition of nitrogen and sulfur is a direct input of potential acidity (ACY), or, in terms of ANC, such deposition is relevant to the major anions that reduce the capacity of a water body to neutralize acidity. Consequently, there is a well defined linear relationship between potential acidifying deposition and ANC. This ANC-deposition relationship facilitates the linkage between ecosystem models that calculate an ecological indicator and the atmospheric deposition of NO_y and SO_x. On the other hand, there is no direct linear relationship between deposition and pH. While acid inputs from deposition lower pH, the relationship can be extremely nonlinear and there is no direct connection from a modeling or mass balance perspective between the amount

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of deposition entering a system and pH. The term "mass balance" underlies the basic formulation of any physical modeling construct, for atmospheric or aquatic systems, and refers to the accounting of the flow of mass into a system, the transformation to other forms, and the loss due to flow out of a system and other removal processes. The ANC is a conserved property. This means that ANC in a water body can be accounted for by knowledge of how much ANC initially exists, how much flows in and is deposited, and how much flows out. In contrast, hydrogen ion concentration in the water, the basis for pH, is not a conserved property as its concentration is affected by several factors such as temperature, atmospheric pressure, mixing conditions of a water body, and the levels of several other chemical species in the system. The disadvantage of pH lacking conservative properties is that there is a very complex connection between changes in ambient air concentrations of NO_y and SO_x and pH.

The discussion of basic water chemistry of natural systems in chapter 2 of the PA provides further details on why pH is not a conserved quantity and is subject to rapid transient response behavior that makes it difficult to use as a reliable and functional ecological indicator. The observed pH-to-ANC relationship (US EPA, 2011, figure 7-2) partially explains the concern with pH responding too abruptly. In the region where pH ranges roughly from 4.5 to 6 and is of greatest relevance to effects (US EPA, 2011, figure 7-4), there clearly is more sensitivity of pH to changes in ANC in the ANC range from approximately 0 to 50 μeq/L. A focus on this part of the ANC-to-pH relationship shows that ANC associates well with pH in a fairly linear manner. However, the pH range from 4.5 to 6 also includes one of the very steepest parts of the slope relating pH as a function of ANC, where ANC ranges down below 0 μeq/L, which is subject to very rapid change in ANC, or deposition inputs. This part of the relationship coincides with reduced levels of ANC and hence with reduced ability to neutralize acids and moderate pH

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fluctuations. This response behavior can be extended to considering how pH would change in response to deposition, or ambient concentrations, of NO_y and SO_x, which can be viewed as "ANC-like" inputs.

In summary, because ANC clearly links both to biological effects of aquatic acidification as well as to acidifying inputs of NO_y and SO_x deposition, the PA concludes that ANC is an appropriate ecological indicator for relating adverse aquatic ecosystem effects to acidifying atmospheric deposition of SO_x and NO_y, and is preferred to other potential indicators. In reaching this conclusion, the PA notes that in its review of the first draft PA, CASAC concluded that "information on levels of ANC protective to fish and other aquatic biota has been well developed and presents probably the lowest level of uncertainty in the entire methodology" (Russell and Samet, 2010a). In its more recent review of the second draft PA, CASAC agreed "that acid neutralizing capacity is an appropriate ecological measure for reflecting the effects of aquatic acidification" (Russell and Samet, 2010b; p. 4).

2. Linking ANC to Deposition

There is evidence to support a quantified relationship between deposition of nitrogen and sulfur and ANC. This relationship was analyzed in the REA for two case study areas, the Adirondack and Shenandoah Mountains, based on time-series modeling and observed trends. In the REA analysis, long-term trends in surface water nitrate, sulfate and ANC were modeled using MAGIC for the two case study areas. These data were used to compare recent surface water conditions in 2006 with preindustrial conditions (i.e. preacidification 1860). The results showed a marked increase in the number of acid impacted lakes, characterized as a decrease in ANC levels, since the onset of anthropogenic nitrogen and sulfur deposition, as discussed in chapter 2 of the PA.

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In the REA, more recent trends in ANC, over the period from 1990 to 2006, were assessed using monitoring data collected at the two case study areas. In both case study areas, nitrate and sulfate deposition decreased over this time period. In the Adirondack Mountains, this corresponded to a decreased concentration of nitrate and sulfate in the surface waters and an increase in ANC (US EPA, 2009, section 4.2.4.2). In the Shenandoah Mountains, there was a slight decrease in nitrate and sulfate concentration in surface waters corresponding to modest increase in ANC from 50 μeq/L in 1990 to 67 μeq/L in 2006 (US EPA, 2009, section 4.2.4.3,Appendix 4, and section 3.4).

In the REA, the quantified relationship between deposition and ANC was investigated using ecosystem acidification models, also referred to as acid balance models or critical loads models (US EPA, 2011, section 2 and US EPA, 2009, section 4 and Appendix 4). These models quantify the relationship between deposition of nitrogen and sulfur and the resulting ANC in surface waters based on an ecosystem's inherent generation of ANC and ability to neutralize nitrogen deposition through biological and physical processes. A critical load is defined as the amount of acidifying atmospheric deposition of nitrogen and sulfur beyond which a target ANC is not reached. Relatively high critical load values imply that an ecosystem can accommodate greater deposition levels than lower critical loads for a specific target ANC level. Ecosystem models that calculate critical loads form the basis for linking deposition to ANC.

As discussed in chapter 2 of the PA, both dynamic and steady state models calculate ANC as a function of ecosystem attributes and atmospheric nitrogen and sulfur deposition, and can be used to calculate critical loads. Steady state models are time invariant and reflect the long term consequences associated with an ecosystem reaching equilibrium under a constant level of atmospheric deposition. Dynamic models are time variant and take into account the

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time dependencies inherent in ecosystem hydrology, soil and biological processes. Dynamic models like MAGIC can provide the time series response of ANC to deposition whereas steady state models provide a single ANC relationship to any fixed deposition level. Dynamic models naturally are more complex than steady state models as they attempt to capture as much of the fundamental biogeochemical processes as practicable, whereas steady state models depend on far greater parameterization and generalization of processes that is afforded, somewhat, by not having to accounting for temporal variability.

The PA notes that steady state models are capable of addressing the question of what does it take to reach and sustain a specific level of ANC. Dynamic models are also capable of addressing that question, but can also address the question of how long it takes to achieve that result. Dynamic models afford the ability for more comprehensive treatment of a variety of processes throughout the surface, soil and bedrock layers within an ecosystem. For example, steady state models treat sulfate as a mobile anion throughout the system, meaning that the sulfate that is deposited to a watershed enters the water column and is not influenced by soil adsorption or cation exchange. Dynamic models can incorporate these time variant processes. The use of a steady state model treating sulfate as totally mobile does not necessarily conflict with the possibility of sulfate acting as a less than mobile ion at certain times. The steady state assumption is premised on the long term behavior of sulfate which can undergo periods of net adsorption followed by periods of net desorption which can balance out over time. The PA recognizes that as the richness of the available data increases, in terms of parameters and spatial resolution, the incorporation of dynamic modeling approaches in the standard setting process should become more feasible. In determining an appropriate modeling approach for the development of a NAAQS in this review, the PA considers both the relevance of the question

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addressed as well as the ability to perform modeling that provides relevant information for geographic areas across the country.

Dynamic models require a large amount of catchment level-specific data relative to steady state models. Because of the time invariant nature of steady state models, the data requirements that integrate across a broad spectrum of ecosystem processes is achievable and available now at the national level. Water quality data to support steady state models currently exist for developing a national data base for modeling nearly 10,000 catchments in the contiguous U.S. In contrast, the data needs to support dynamic models for national-scale analyses simply are not available at this time. Further, the information provided by steady state modeling would be sufficient to develop and analyze alternative NAAQS and the kind of protection they would afford. While it would be of interest to also obtain information about how much time it would take for a target ANC level to be achieved, the absence of such information does not preclude developing and evaluating alternative NAAQS using the AAI structure. Based on the above considerations, the PA concludes that at this time steady state critical load modeling is an appropriate tool for linking long-term ANC levels to atmospheric deposition of nitrogen and sulfur for development of an AAI that has national applicability.

A steady state model is used to define the critical load, which is the amount of atmospheric deposition of nitrogen (N) and sulfur (S) beyond which a target ANC is not achieved and sustained.[7] It is expressed as:

$$CL_{ANClim}(N + S) = ([BC]_0^* - [ANC_{lim}])Q + Neco \qquad \text{(III-1)}$$

[7] This section discusses the linkages between deposition of nitrogen and sulfur and ANC. Section III.B.3 then discusses the linkages between atmospheric concentrations of NOy and SOx and deposition of nitrogen and sulfur.

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Where:

$CL_{ANClim}(N + S)$ is the critical load of deposition, with units of equivalent charge/(area-time);

$[BC]_0^*$ is the natural contribution of base cations from weathering, soil processes and preindustrial deposition, with units of equivalent charge/volume;

$[ANC_{lim}]$ is the target ANC value, with units of equivalent charge/volume;

Q is the catchment level runoff rate governed by water mass balance and dominated by precipitation, with units of distance/time; and

Neco is the amount of nitrogen deposition that is effectively neutralized by a variety of biological (e.g., nutrient uptake) and physical processes, with units of equivalent charge/ (area-time).

Equation III-1 is a modified expression that adopts the basic formulation of the steady state models that are described in chapter 2 of the PA. More detailed discussion of the rationale, assumptions and derivation of equation III-1, as well as all of the equations in this section, are included in Appendix B of the PA. The equation simply reflects the amount of deposition of nitrogen and sulfur from the atmosphere, $CL_{ANClim}(N + S)$, that is associated with a sustainable long-term ANC target, $[ANC_{lim}]$, given the capacity of the natural system to generate ANC, $[BC]_0^*$, and the capacity of the natural system to neutralize nitrogen deposition, *Neco*. This expression of critical load is valid when nitrogen deposition is greater than *Neco*.[8] The runoff rate, *Q*, allows for balancing mass in the two environmental mediums – atmosphere and catchment. This critical load expression can be focused on a single water system or more broadly. To extend applicability of the critical load expression (equation III-1) from the

[8] Because *Neco* is only relevant to nitrogen deposition, in rare cases where *Neco* is greater than the total nitrogen deposition, the critical load would be defined only in terms of acidifying deposition of sulfur and the *Neco* term in equation III-1 would be set to zero.

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catchment level to broader spatial areas, the terms Q_r and CL_r, are used, which are the runoff rate and critical load, respectively, of the region over which all the atmospheric terms in the equation are defined.

In considering the contributions of SO_x or NO_y species to acidification, it is useful to think of every depositing nitrogen atom as supplying one equivalent charge unit and every sulfur atom as depositing two charge units. The PA uses equivalent charge per volume as a normalizing tool in place of the more familiar metrics such as mass or moles per volume. This allows for a clearer explanation of many of the relationships between atmospheric and ecosystem processes that incorporate mass and volume unit conventions somewhat specific to the environmental media of concern (e.g., m^3 for air and liter for liquid water). Equivalent charge reflects the chemistry equilibrium fundamentals that assume electroneutrality, or balancing charge where the sum of cations always equals the sum of anions.

As presented above, the terms S and N in the CL_{ANClim} $(N + S)$ term broadly represent all species of sulfur or nitrogen that can contribute to acidifying deposition. This follows conventions used in the scientific literature that addresses critical loads, and it reflects all possible acidifying contributions from any sulfur or nitrogen species. For all practical purposes, S reflects SOx as described above, the sum of sulfur dioxide gas and particulate sulfate. However, N in equation III-1 includes both oxidized forms, consistent with the ambient indicator, NO_y, in addition to the reduced nitrogen species, ammonia and ammonium ion, referred to as NHx. The NH_x is included in the critical load formulation because it contributes to potentially acidifying nitrogen deposition. Consequently, from a mass balance or modeling perspective, the form of the standard needs to account for NH_x, as described below.

3. Linking Deposition to Ambient Air Indicators

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The last major component of the form illustrated in Figure III-1 addresses the linkage between deposition of nitrogen and sulfur and concentrations of the ambient air indicators, NO_y and SO_x. To link ambient air concentrations with deposition, the PA defines a transference ratio, T, as the ratio of total wet and dry deposition to ambient concentration, consistent with the area and time period over which the standard is defined. To express deposition of NO_y and SO_x in terms of NO_y and SO_x ambient concentrations, two transference ratios were defined, where T_{SOx} equals the ratio of the combined dry and wet deposition of SO_x to the ambient air concentration of SO_x, and T_{NOy} equals the ratio of the combined dry and wet deposition of NO_y to the ambient air concentration of NO_y.

As described in chapter 7 of the PA, reduced forms of nitrogen (NH_x) are included in total nitrogen in the critical load equation, III-1. Reduced forms of nitrogen are treated separately, as are NO_y and SO_x, and the transference ratios are applied. This results in the following critical load expression that is defined explicitly in terms of the indicators NO_y and SO_x:

$$CL_{ANClim}(N + S) = ([BC]_0^* - [ANC_{lim}])Q + Neco = [NOy]T_{NOy} + [SOx]T_{SOx} + NHx \qquad \text{(III-2)}$$

This is the same equation as III-1, with the deposition associated with the critical load translated to deposition from ambient air concentrations via transference ratios. In addition, deposition of reduced nitrogen, oxidized nitrogen and oxidized sulfur are treated separately.

Transference ratios are a modeled construct, and therefore cannot be compared directly to measurable quantities. There is an analogy to deposition velocity, as a transference ratio is basically an aggregated weighted average of the deposition velocities of all contributing species

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across dry and wet deposition, and transference ratio units are expressed as distance/time. However, wet deposition commonly is not interpreted as the product of a concentration times a velocity. Direct wet deposition observations are available which integrate all of the processes, regardless of how well they may be understood, related to wet deposition into a measurable quantity. There are reasonable analogies between the processes governing dry and wet deposition, from a fundamental mass transfer perspective. In both cases there is a transfer of mass between the dry ambient phase and another medium, either a surface or vegetation in the case of dry deposition, or a rain droplet or cloud in the case of wet precipitation. The specific thermodynamic properties and chemical/biological reactions that govern the transfer of dry mass to plants or aqueous droplets differ, but either process can be based on conceptualizing the product of a concentration, or concentration difference, times a mass transfer coefficient which is analogous to the basic dry deposition model: dry deposition = concentration x velocity (US EPA, 2011, Appendix F).

Transference ratios require estimates of wet deposition of NO_y and SO_x, dry deposition of NO_y and SO_x, and ambient air concentrations of NO_y and SO_x. Possible sources of information include model estimates or a combination of model estimates and observations, recognizing that dry deposition is a modeled quantity that can use observed or modeled estimates of concentration. The limited amount of NO_y measurements in acid-sensitive areas as well as the combination of representative NO_y, SO_2 and SO_4 observations generally preclude the use of observations for development of a standard that is applicable nationally.

The PA considers a blending of observations and models to take advantage of their relative strengths; e.g., combining the NADP wet deposition observations, modeled dry deposition, and a mix of modeled and observed concentrations, using the model for those species

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not measured or measured with very sparse spatial coverage. A potential disadvantage of mixing and matching observations and model estimates is to lose consistency afforded by using just modeling alone. A modeling platform like CMAQ is based on adhering to consistent treatment of mass conservation, by linking emission inputs with air concentrations and concentrations to deposition. Inconsistencies from combining processes from different analytical platforms increase the chance that mass (of nitrogen or sulfur) would unintentionally be increased or decreased as the internal checking that assures mass conservation is lost. Transference ratios incorporate a broad suite of atmospheric processes and consequently an analytical approach that instills consistency in the linkage of these processes is preferable to an approach lacking such inherent consistency. This contention does not mean that observations alone, if available, could not be used, but suggests that the inconsistencies in combining models and observations for the purposes of developing transference ratios has the potential for creating unintended artifacts.

While there is a reasonable conceptual basis for the concept of an aggregated deposition velocity referred to in the PA as a transference ratio, there is very limited ability to compare observed and calculated ratios. This is because the deposition velocity is dependent on individual species, and the mass transfer processes of wet and dry removal, while conceptually similar, are different. Consequently, there does not exist a meaningful approach to measure such an aggregated or lumped parameter. Therefore, at this time, the evaluation of transference ratios is based on sensitivity studies, analysis of variability, and comparisons with other models, as described in Appendix F of the PA.

As discussed in Appendix F, the interannual variability, as well as the sensitivity to emission changes of roughly 50 percent, results in changes of transference ratios of approximately 5 to 10 percent. Part of the reason for this inherent stability is due to the co-

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dependence of concentration and deposition. For example, as concentrations are reduced as a result of emissions reductions, deposition in turn is reduced since deposition is a direct linear function of concentration leading to negligible impact on the deposition-to-concentration ratio. Likewise, an overestimate of concentration likely does not induce a bias in the transference ratio. While it is important to continue to improve the model's ability to match ambient concentrations in time and space, the bias of a modeled estimate of concentration relative to observations does not necessarily result in a bias in a calculated transference ratio. In effect, this consideration of bias cancellation reduces the sensitivity of transference ratios to model uncertainties and affords increased confidence in the stability of these ratios. Based on the series of sensitivity and variability analyses, the PA concludes that the transference ratios are relatively stable and provide a sound metric for linking deposition and concentration.

As discussed in the PA, transference ratios are dependent on the platform upon which they are constructed. Comparisons of transference ratios constructed from different modeling platforms do exhibit significant differences. While this divergence of results may be explained by a variety of differences in process treatments, input fields and incommensurabilities in species definitions and spatial configurations, it does suggest two very important conclusions. First, the idea of using multiple platforms for different parts of the country may be problematic as there does not exist a reliable approach to judge acceptance which is almost always based on comparisons to observations. Second, since transference ratios are based on concentrations and deposition, as the uncertainties in each of those components are reduced, the relative uncertainty in the ratios also is reduced. This means that basic improvements in the model's ability to reproduce observed wet deposition and ambient concentration fields enhance the relative confidence in the constructed transference ratios. Similarly, as in-situ dry deposition flux

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measurements become available that enable a more rigorous evaluation and diagnosis of modeled dry deposition processes, the expected improved treatment of dry deposition also would increase confidence in transference ratios. Finally, deposition is directly related to ambient air concentrations. Models like CMAQ rely on the concentration-to-deposition linkage to calculate deposition, which is the foundation for broadly based and robust assessments addressing atmospheric deposition. In principle, the use of a modeled constructed transference ratio is based on the same premise by which we use models to estimate deposition in the first place.

The shortage of widely available ambient air observations and the fact that estimates of dry deposition requires modeling, collectively suggests that a unified modeling platform is the best approach for constructing transference ratios. The PA (US EPA, 2011, section 2) considers CMAQ and other models, such as CAMx and Canada's AURAMS - A Unified Regional Air-quality Modeling System (Smythe et al., 2008), and concludes that CMAQ is the preferred modeling platform for constructing transference ratios. This conclusion reflects the view that for the purposes of defining transference ratios, a modeling platform should: (1) be a multiple pollutant model recognizing the myriad of connections across pollutant categories that directly and indirectly impact nitrogen and sulfur characterization, (2) include the most comprehensive scientific treatments of atmospheric processes that relate directly and indirectly to characterizing concentrations and deposition, (3) have an infrastructure capability that accommodates the inclusion of improved scientific treatments of relevant processes and important input fields, and (4) undergo frequent reviews regarding the adequacy of the underlying science as well as the appropriateness in applications. The CMAQ platform exhibits all these characteristics. It has been (and continues to be) extensively evaluated for several pollutant categories, and is supported by a central infrastructure of EPA scientists, whose mission is to improve and evaluate

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the CMAQ platform. More directly, CMAQ, and its predecessor versions, has a long track record going back to the NAPAP in the 1980's of specific improvements in deposition processes, which are described in Appendix F of the PA.

4. Aquatic Acidification Index

Having established the various expressions that link atmospheric deposition of nitrogen and sulfur to ANC and the transference ratios that translate atmospheric concentrations to deposition of nitrogen and sulfur, the PA derived the following expression of these linkages, which separates reduced forms of nitrogen, NHx, from oxidized forms:

$$ANCcalc = \{ANClim + CL_r/Q_r\} - NHx/Q_r - T_{NOy}[NOy]/Q_r - T_{SOx}[SOx]/Q_r \qquad \text{(III-3)}$$

Equation III-3 is the basic expression of the form of a standard that translates the conceptual framework into an explicit expression that defines ANC as a function of the ambient air indicators, NO_y and SO_x reduced nitrogen deposition[9], and the critical load necessary to achieve a target ANC level. This equation calculates an expected ANC value based on ambient concentrations of NO_y and SO_x. The calculated ANC will differ from the target ANC (ANClim) depending on how much the nitrogen and sulfur deposition associated with NO_y, SO_x, and NH_x differs from the critical load associated with just achieving the target ANC.

Based on equation III-3, the PA defines an AAI that is more simply stated using terms that highlight the ambient air indicators:

[9] Because NHx is characterized directly as deposition, not as an ambient concentration in this equation, no transference ratio is needed for this term.

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$$AAI = F1 - F2 - F3[NOy] - F4[SOx] \quad \text{(III- 4)}$$

where the AAI represents the long term (or steady state) ANC level associated with ambient air concentrations of NO_y and SO_z. The factors F1 through F4 convey three attributes: a relative measure of the ecosystem's ability to neutralize acids (F1), the acidifying potential of reduced nitrogen deposition (F2), and the deposition-to-concentration translators for NOy (F3) and SOx (F4).

Specifically:

F1 = ANClim + CL_r/Q_r ;

F2 = NHx/Q_r = NHx deposition divided by Q_r;

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

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

All of these factors include representative Q_r to maintain unit (and mass) consistency between the AAI and the terms on the right side of equation III-4.

The F1 factor is the target ANC level plus the amount of deposition (critical load) the ecosystem can receive and still achieve the target level. It incorporates an ecosystem's ability to generate acid neutralizing capacity through base cation supply *([BC]*$_0$)* and to neutralize acidifying nitrogen deposition through *Neco,* both of which are incorporated in the CL term. As noted above, because *Neco* can only neutralize nitrogen deposition (oxidized or reduced) there may be rare cases where *Neco* exceeds the combination of reduced and oxidized nitrogen deposition. Consequently, to ensure that the AAI equation is applicable in all cases that may

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occur, equation III-4 is conditional on total nitrogen deposition, {NHx + F3[NOy]}, being greater than *Neco*. In rare cases where *Neco* is greater than {NHx + F3[NOy]}, F2, F3, and Neco would be set equal to 0 in the AAI equation. The consequence of setting F2 and F3 to zero is simply to constrain the AAI calculation just to SO_x, as nitrogen would have no bearing on acidifying contributions in this case.

The PA concludes that equation III-4 (US EPA, 2011,equation 7-12), which defines the AAI, is ecologically relevant and appropriate for use as the form of a national standard designed to provide protection for aquatic ecosystems from the effects of acidifying deposition associated with concentrations of oxides of nitrogen and sulfur in the ambient air. This AAI equation does not, however, in itself, define the spatial areas over which the terms of the equation would apply. To specify values for factors F1 through F4, it is necessary to define spatial areas over which these factors are determined. Thus, it is necessary to identify an approach for spatially aggregating water bodies into ecologically meaningful regions across the U.S., as discussed below.

5. Spatial Aggregation

As discussed in the PA, one of the unique aspects of this form is the need to consider the spatial areas over which values for the F factors in the AAI equation are quantified. Ecosystems across the U.S. exhibit a wide range of geological, hydrological and vegetation characteristics that influence greatly the ecosystem parameters, Q, BC_0^* and *Neco* that are incorporated in the AAI. Variations in ecosystem attributes naturally lead to wide variability in the sensitivities of water bodies in the U.S. to acidification, as well as in the responsiveness of water bodies to changes in acidifying deposition. Consequently, variations in ecosystem sensitivity, and the uncertainties inherent in characterizing these variations, must be taken into account in

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developing a national standard. In developing a secondary NAAQS to protect public welfare, the focus of the PA is on protecting sensitive populations of water bodies, not on each individual water body, which is consistent with the Agency's approach to protecting public health through primary NAAQS that focus on susceptible populations, not on each individual.

The approach used for defining ecologically relevant regions across the U.S. in the PA (US EPA, 2011, section 7.2.5) is described below, along with approaches to characterizing each region as acid sensitive or relatively non-acid sensitive. This characterization facilitates a more detailed analysis and focus on those regions that are relatively more acid sensitive. This characterization is also used to avoid over-protection in relatively non-acid sensitive regions, regions that would receive limited benefit from reductions in the deposition of oxides of nitrogen and sulfur with respect to aquatic acidification effects. Approaches to developing representative values for each of the terms in the AAI equation (factors F1 through F4) for each ecologically relevant region for which sufficient data are available are then discussed. These spatial aggregation approaches are generally applicable to the contiguous U.S.. The following discussion also addresses the development of factors for data-limited regions and specifically for Hawaii, Alaska and the U.S. territories.

Stated more simply, this section discusses appropriate ways to divide the country into ecologically relevant regions; to characterize each region as either acid sensitive or relatively non-acid sensitive; and to determine values of factors F1 through F4 for each region, taking into consideration the acid sensitivity of each region and the availability of relevant data. For each such region, the AAI would be calculated based on the values of factors F1 through F4 specified for that region.

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In considering approaches to spatial aggregation, the PA focuses on methods that have been developed to define ecologically relevant regions, referred to as ecoregions, which are meaningfully related to the factors that are relevant to aquatic acidification. As noted above, the PA did not focus on looking at each individual water body, nor did it focus on aggregating over the entire nation, which would preclude taking into account the inherent variability in atmospheric and ecological factors that fundamentally modify the relationships that are central to the development of an ecologically relevant AAI.

Based on considering available classification schemes, the PA concludes that Omernik's ecoregion classification (as described at http://www.epa.gov/wed/pages/ecoregions) is the most appropriate method to consider for the purposes of this review. This classification offers several levels of spatial delineation, has undergone an extensive scientific peer review process, and has explicitly been applied to delineating acid sensitive areas within the U.S. Further, the PA concludes that ecoregion level III (Figure III-1) resolution, with 84 defined ecoregions in the contiguous U.S.,[10] is the most appropriate level to consider for this purpose. The spatial resolution afforded by level III strikes an appropriate balance relative to the reasoning that supports conclusions on indicators, as discussed above. The PA concludes that the most detailed level of resolution (level IV) is not appropriate given the limited data availability to address nearly 1000 subdivisons within that level and the currently evolving nature of level IV regions. Further, level III ecoregions are preferred to level II in that level III ecoregions, but not level II ecoregions, are largely contiguous in space which allows for a more coherent development of information to quantify the AAI factors and to characterize the concentrations of NOy and SOx in the ambient air within each ecoregion.

[10] We note that an 85th area within Omernik's Ecoregion Level III is currently being developed for California.

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Appendix C of the PA includes a description of each level III ecoregion. The PA notes that the use of ecoregions is an appropriate spatial aggregation scheme for an AAI-based standard focused on deposition-related aquatic acidification effects, while many of the same ecoregion attributes may be applicable in subsequent NAAQS reviews that may address other deposition-related aquatic and terrestrial ecological effects. Because atmospheric deposition is modified by ecosystem attributes, the types of vegetation, soils, bedrock geology, and topographic features that are the basis of this ecoregion classification approach also will likely be key attributes for other deposition-related effects (e.g., terrestrial acidification, nutrient enrichment) that link atmospheric concentrations to an aquatic or terrestrial ecological indicator.

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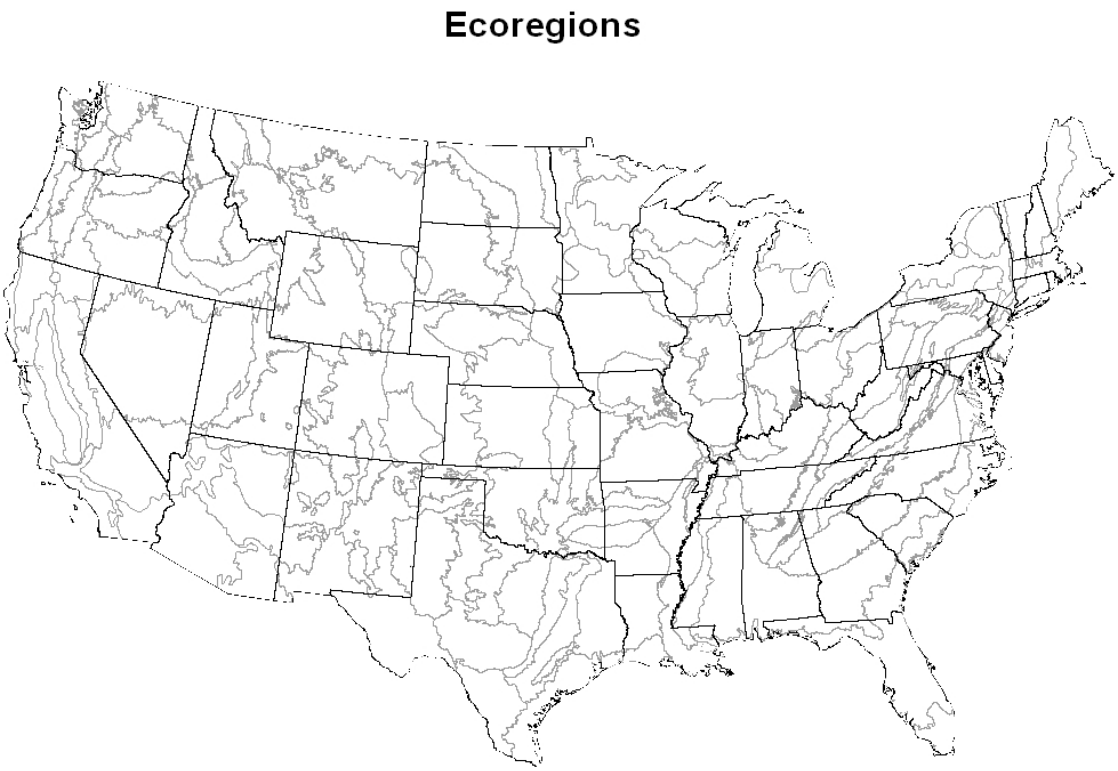

Figure III-1. Omernik Ecoregion III areas (http://www.epa.gov/wed/pages/ecoregions).

a. Ecoregion Sensitivity

The PA used Omernik's original alkalinity data (US EPA, 2011, section 2) and more recent ANC data to delineate two broad groupings of ecoregions: acid-sensitive and relatively non-acid sensitive ecoregions. This delineation was made to facilitate greater focus on those ecoregions with water bodies that generally have greater acid sensitivity and to avoid over-protection in regions with generally less sensitive water bodies. The approach used to delineate acid-sensitive and relatively non-acid sensitive regions included an initial numerical-based sorting scheme using ANC data, which categorized ecoregions with relatively high ANC values

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as being relatively non-acid sensitive. This initial delineation resulted in 29 of the 84 Omernik ecoregions being categorized as acid sensitive. Subsequently, land use data were also considered to determine to what extent an ecoregion is of a relatively pristine and rural nature by quantifying the degree to which active management practices related to development and agriculture occur in each ecoregion.

The overall objective is to produce a logical and practical grouping of ecoregions that experience adverse conditions with respect to aquatic acidification and are likely to respond to changes in concentrations of NO_y and SO_x in the ambient air and to the related deposition levels. To achieve this goal, a two-step process has been applied, first identifying acid sensitive ecoregions based on water quality data alone, and second identifying among those acid-sensitive ecoregions those with highly developed and managed areas. These highly developed and managed ecoregions are placed in a non-acid sensitive category to avoid over protection beyond what is requisite to protect public welfare. More specifically, in determining an ecoregion's acid sensitivity status in step 1, ANC data across the 84 ecoregions are sorted (US EPA, 2011, section 7) to determine the number of water bodies within a region with ANC values suggestive of acid sensitivity, so as to screen out regions with an overabundance of high ANC values. In reviewing the ANC data, the PA identified 29 ecoregions that meet two criteria: (1) greater than 5 percent of water bodies with data with ANC values less than 200 μeq/L and (2) greater than 1 percent of water bodies with ANC values less than 100 μeq/L. In step 2, land use data were used to identify those acid sensitive ecoregions with significant managed areas that would not be considered as having a relatively pristine and rural character. The percentage of the combination of developed (residential, transportation, industrial and commercial) and agricultural (croplands, pastures, orchards, vineyards) land use was used as an indicator of managed land use area. Forest cover

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was used as an indicator of non-managed land use more directly reflecting the pristine quality of a region. Based on the 2006 National Land Cover Data base (NLCD, http://www.epa.gov/mrlc/nlcd-2006.html), acid sensitive ecoregions would meet both of the following land use data criteria: percent of developed and agricultural area less than 20 percent combined with forested area greater than 50 percent. The combination of steps 1 and 2 identify 22 relatively acid sensitive areas (Table III-1 and Figure III-2).

Consideration was also given to the use of naturally acidic conditions in defining relatively non-acid sensitive areas. For example, several of the ecoregions located in plains near the coast exhibit elevated dissolved organic carbon (DOC) levels, which is associated with naturally acidic conditions. The DOC in surface waters is derived from a variety of weak organic acid compounds generated from the natural availability and decomposition of organic matter from biota. Consequently, high DOC is associated with "natural" acidity, with the implication that a standard intended to protect against atmospheric contributions to acidity is not an area of focus. The evidence suggests that several of the more highly managed ecoregions in coastal or near coastal transition zones are associated with relatively high DOC values, typically exceeding on average 5 mg/l, compared to other acid sensitive areas. Although there is sound logic to interpret naturally acidic areas as relatively non-acid sensitive, natural acidity indicators were not explicitly included in defining relatively non-acid sensitive areas as there does not exist a consensus-based quantifiable scientific definition of natural acidity. Approaches to explicitly define natural acidity likely will be pursued in future reviews of the standard.

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Table III-1. List of 22 Acid-Sensitive Areas	
Ecoregion name	**Ecoregion number**
Ridge and Valley	8.4.1
Northern Appalachian Plateau and Uplands	8.1.3
Piedmont	8.3.4
Western Allegheny Plateau	8.4.3
Southwestern Appalachians	8.4.9
Boston Mountains	8.4.6
Blue Ridge	8.4.4
Ouachita Mountains	8.4.8
Central Appalachians	8.4.2
Northern Lakes and Forests	5.2.1
Maine/New Brunswick Plains and Hills	8.1.8
North Central Appalachians	5.3.3
Northern Appalachian and Atlantic Maritime Highlands	5.3.1
Columbia Mountains/Northern Rockies	6.2.3
Middle Rockies	6.2.10
Wasatch and Uinta Mountains	6.2.13
North Cascades	6.2.5
Cascades	6.2.7
Southern Rockies	6.2.14
Sierra Nevada	6.2.12
Idaho Batholith	6.2.15
Canadian Rockies	6.2.4

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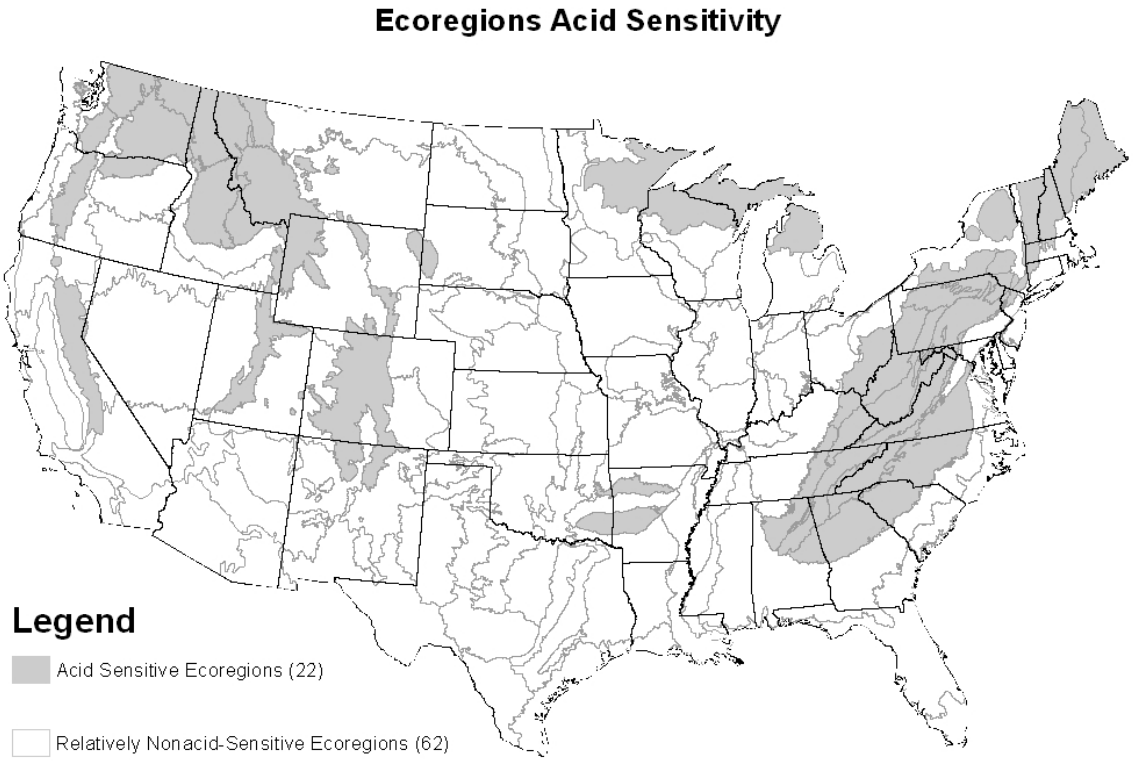

Figure III-2. Acid-sensitive ecoregions identified in grey fill (22 out of 84 ecoregions).

b. Representative Ecoregion-specific Factors

Having concluded that the Omernik level III ecoregions are an appropriate approach to spatial aggregation for the purpose of a standard to address deposition-related aquatic acidification effects, the PA uses those ecoregions to define each of the factors in the AAI equation. As discussed below, factors F1 through F4 in equation III-4 are defined for each ecoregion by specifying ecoregion-specific values for each factor based on monitored or modeled data that are representative of each ecoregion.

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i. Factor F1

As discussed above, factor F1 reflects a relative measure of an ecosystem's ability to neutralize acidifying deposition, and is defined as: F1 = ANClim + CL_r/Q_r. The value of F1 for each ecoregion would be based on a representative critical load for the ecoregion (CL_r) associated with a single national target ANC level (ANClim, discussed below in section III.D), as well as on a representative runoff rate (Q_r). To specify ecoregion-specific values for the term Q_r, the PA used the median value of the distribution of Q values that are available for water bodies within each ecoregion. To specify ecoregion-specific representative values for the term CL_r in factor F1, a distribution[11] of calculated critical loads was created for the water bodies in each ecoregion for which sufficient water quality and hydrology data are available.[12] The representative critical load was then defined to be a specific percentile of the distribution of critical loads in the ecoregion. Thus, for example, using the 90th percentile means that within an ecoregion, 90 percent of the water bodies would be expected to have higher calculated critical loads than the representative critical load. That is, if the representative critical load were to occur across the ecoregion, 90 percent of the water bodies would be expected to achieve the national ANC target or better.

The specific percentile selected as part of the definition of F1 is an important parameter that directly impacts the representative critical load specified for each ecoregion, and therefore the degree of protectiveness of the standard. A higher percentile corresponds to a lower critical load and, therefore, to lower allowable ambient air concentrations of NO_y and SO_x and related

[11] The distribution of critical loads was based on CL values calculated with Neco at the lake level. Consideration could also be given to using a distribution of CLs without Neco and adding the ecoregion average Neco value to the nth percentile critical load. This would avoid cases where the lake-level Neco value potentially could be greater than total nitrogen deposition. The CL at the lake level represents the CL for the lake to achieve the specified national target ANC value.

[12] The PA judged the data to be sufficient for this purpose if data are available from more than 10 water bodies in an ecoregion.

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deposition to achieve a target AAI level. In conjunction with the other terms in the AAI equation, alternative forms can be appropriately characterized in part by identifying a range of alternative percentiles. The choice of an appropriate range of percentiles to consider for acid-sensitive and relatively non-acid sensitive ecoregions, respectively, is discussed below.

(a) Acid-sensitive Ecoregions

In identifying percentiles that are appropriate to consider for the purpose of calculating factor F1 for ecoregions characterized as acid sensitive, the PA concludes that it is appropriate to focus on the lower (more sensitive) part of the distribution of critical loads, so as to ensure that the ecoregion would be represented by relatively more acid sensitive water bodies within the ecoregion. Specifying factor F1 in this way would help to define a standard that would be protective of the population of acid sensitive water bodies within an ecoregion, recognizing that even ecoregions characterized as acid sensitive may contain a number of individual water bodies that are not acid sensitive. The PA recognizes that there is no basis for independently evaluating the degree of protectiveness afforded by any specific percentile value, since it is the combination of form and level, in conjunction with the indicator and averaging time, which determine the degree of protectiveness of a standard. In light of this, the PA concludes that it is appropriate to consider a range of percentiles, from well above the 50th percentile, or median, of the distribution to somewhat below the highest value (in terms of sensitivity; a high degree of sensitivity corresponds to a low value for critical load). More specifically, the PA concludes it is appropriate to consider percentiles in the range of the 70th to the 90th percentile (of sensitivity). This conclusion is based on the judgment that it would not be appropriate to represent an ecoregion with the lowest or near lowest critical load, so as to avoid potential extreme outliers that can be seen to exist at the extreme end of the data distributions, which would not be

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representative of the population of acid sensitive water bodies within the ecoregion and could lead to an overly protective standard. At the same time, in considering ecoregions that are inherently acid sensitive, it is judged to be appropriate to limit the lower end of the range for consideration to the 70th percentile, a value well above the median of the distribution, so that a substantial majority of acid-sensitive water bodies are protected.

In considering this conclusion, the CASAC Panel noted that the data bases for calculating critical loads within an ecoregion are not necessarily representative of all water bodies within an ecoregion. That is, in many ecoregions the lake sampling design used in studies that generated the relevant data may have focused on the relatively more sensitive water bodies within an ecoregion (Russell and Samet, 2011a). Consequently, a given percentile of the distribution of calculated critical loads, based on sampled water bodies, may not be representative of that percentile of all water bodies across an entire ecoregion. To the extent that the sampling of water bodies within an ecoregion was skewed toward the relatively more sensitive water bodies, selecting a given percentile from the distribution of available critical loads would result in a somewhat higher percentile of all water bodies within that ecoregion having a higher calculated critical load than the representative critical load value. Thus, the extent to which study sampling designs have resulted in skewed distributions of calculated critical loads is an uncertainty that is appropriate to consider in selecting a percentile for the purpose of defining the factor F1 in the AAI equation.

(b) Non-acid sensitive Ecoregions

With regard to identifying percentiles that are appropriate to consider for the purpose of calculating factor F1 for ecoregions characterized as relatively non-acid sensitive, the PA recognizes that while such ecoregions are generally less sensitive to acidifying deposition from

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oxides of nitrogen and sulfur, they may contain a number of water bodies that are acid sensitive This category includes ecoregions that are well protected from acidification effects due to natural production of base cations and high ANC levels, as well as naturally acidic systems with limited base cation production and consequently very low critical loads. Therefore, the use of a critical load that would be associated with a highly sensitive water body in a naturally acidic system would impose a high degree of relative protection in terms of allowable ambient air concentrations of oxides of nitrogen and sulfur and related deposition, while potentially affording little or no public welfare benefit from attempting to improve a naturally acidic system.

Based on these considerations, the PA concludes it is appropriate to consider the use of a range of percentiles that extends lower than the range identified above for acid-sensitive ecoregions. Consideration of a lower percentile would avoid representing a relatively non-acid sensitive ecoregion by a critical load associated with relatively more acid-sensitive water bodies. In particular, the PA concludes it is appropriate to focus on the median or 50th percentile of the distribution of critical loads so as to avoid over-protection in such ecoregions. Recognizing that relatively non-acid sensitive ecoregions generally are not sampled to the extent that acid-sensitive ecoregions are, it also is appropriate to consider using the median critical load of all relatively non-acid sensitive ecoregions for each such ecoregion.

ii. Factor F2

As discussed above, factor F2 is the amount of reduced nitrogen deposition within an ecoregion, including the deposition of both ammonia gas and ammonium ion, and is defined as: $F2 = NHx/Q_r$. The PA calculated the representative runoff rate, Q_r, using a similar approach as noted above for factor F1; i.e., the median value of the distribution of Q values that are available for water bodies within each ecoregion. In the PA, 2005 CMAQ model simulations over 12-km

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grids are used to calculate an average value of NHx for each ecoregion. The NHx term is based on annual average model outputs for each grid cell, which are spatially averaged across all the grid cells contained in each ecoregion to calculate a representative annual average value for each ecoregion. The PA concludes that this approach of using spatially averaged values is appropriate for modeling, largely due to the relatively rapid mixing of air masses that typically results in relatively homogeneous air quality patterns for regionally dispersed pollutants. In addition, there is greater confidence in using spatially averaged modeled atmospheric fields than in using modeled point-specific fields.

This averaging approach is also used for the air concentration and deposition terms in factors F3 and F4, as discussed below. The PA notes that modeled NH_x deposition exhibits greater spatial variability than the other modeled terms in factors F3 and F4. Recognizing this greater variability, the PA concludes that it would be appropriate to consider alternative approaches to specifying the value of NH_x. One such approach might involve the use of more localized and/or contemporaneous modeling in areas where this term is likely to be particularly variable and important.

iii. Factors F3 and F4

As discussed above, factors F3 and F4 are the ratios that relate ambient air concentrations of NOy and SOx to the associated deposition, and are defined as follow: F3 = T_{NOy}/ Q_r and F4 = T_{SOx}/ Q_r. T_{NOy} is the transference ratio that converts ambient air concentrations of NO_y to deposition of NO_y and T_{SOx} is the transference ratio that converts ambient air concentrations of SOx to deposition of SOx. The representative runoff rate, Q_r, is calculated as for factors F1 and F2. The transference ratios are based on the 2005 CMAQ simulations, using average values for each ecoregion, as noted above for factor F2. More specifically, the transference ratios are

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calculated as the annual deposition of NO_y or SO_x spatially averaged across the ecoregion and divided by the annual ambient air concentration of NO_y or SO_x, respectively, spatially averaged across the ecoregion.

c. Factors in Data-limited Ecoregions

As discussed above in section III.B.5.a, in the PA the initial delineation of acid-sensitive and relatively non-acid sensitive ecoregions was based on available ANC and alkalinity data. Areas not meeting the ANC criteria described above are categorized as relatively non-acid sensitive. The development of a reasonable distribution of critical loads for water bodies within an ecoregion for the purpose of identifying the representative critical load requires additional data, including more specific water quality data for major cations and anions. This means that the water bodies that can be used to develop a distribution of critical loads is generally a subset of those water bodies for which ANC data are available Consequently, there are certain ecoregions with sparse data that are not suitable for developing a distribution of critical loads.

As noted above, the PA judges that it is not appropriate to develop such distributions based on data from less than ten water bodies within an ecoregion. Twelve such ecoregions, which included only relatively non-acid sensitive ecoregions, were characterized as being data-limited. For these ecoregions, the PA considered alternative approaches to specifying values for the terms CL_r and Q_r for the purpose of determining values for each of the factors in the AAI equation. For these data-limited ecoregions, the PA judges that it is appropriate to use the median values of CL_r and Q_r from the distributions of these terms for all other relatively non-acid sensitive ecoregions, rather than attempting to use severely limited data to develop a value for these terms based solely on data from such an ecoregion. Further, consideration could be given to using a single national default value for all relatively non-acid sensitive ecoregions. The PA

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notes that this data limitation is not a concern in specifying values for the other terms in the AAI equation for such ecoregions, since those terms are based on data from the 2005 CMAQ model simulation, which covers all ecoregions across the contiguous U.S.

d. Application to Hawaii, Alaska, and the U.S. Territories

The above methods for specifying ecoregion-specific values for the factors in the AAI equation apply to those ecoregions within the contiguous U.S. For areas outside the continental U.S., including Hawaii, Alaska, and the U.S. Territories, there is currently a lack of available data to characterize the sensitivity of such areas, as well as a lack of water body-specific data and CMAQ-type modeling to specify values for the F1 through F4 factors. Thus, the PA has considered possible alternative approaches to specifying values for factors F1 through F4 in the AAI equation for these areas.

One such approach could be to specify area-specific values for the factors based on values derived for ecoregions with similar acid sensitivities, to the extent that relevant information can be obtained to determine such similarities. Such an approach would involve conducting an analysis to characterize similarities in relevant ecological attributes between ecoregions in the contiguous U.S. and these areas outside the contiguous U.S. so as to determine the appropriateness of utilizing ecoregion-specific values for the CL_r and Q_r terms from one or more ecoregions within the contiguous U.S. This approach would also involve conducting additional air quality modeling for the areas that are outside the geographical scope of the currently available CMAQ model simulations, so as to develop the other information necessary to specify values for factors F2 through F4 for these areas.

A second approach could rely on future data collection efforts to establish relevant ecological data within these areas that, together with additional air quality modeling, could be

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used to specify area-specific values for factors F1 through F4. Until such time as relevant data become available, these areas could be treated the same as data-limited ecoregions in the contiguous U.S. that are relatively non-acid sensitive.

The PA concludes that either approach would introduce substantial uncertainties that arise from attempting to extrapolate values based on similarity assumptions or arbitrarily assigning values for factors in the AAI equation that would be applicable to these areas outside the contiguous U.S. In light of such uncertainties, the PA concludes that it would also be appropriate to consider relying on the existing NO_2 and SO_2 secondary standards in these areas for protection of any potential direct or deposition-related ecological effects that may be associated with the presence of oxides of nitrogen and sulfur in the ambient air. The PA concludes that relying on existing secondary standards in these areas is preferable to using a highly uncertain approach to allow for the application of a new standard based on the AAI in the absence of relevant area-specific data.

6. Summary of the AAI Form

With regard to the form of a multi-pollutant air quality standard to address deposition-related aquatic acidification effects, the PA concludes that consideration should be given to an ecologically relevant form that characterizes the relationships between the ambient air indicators for oxides of nitrogen and sulfur, the related deposition of nitrogen and sulfur, and the associated aquatic acidification effects in terms of a relevant ecological indicator. Based on the available information and assessments, consideration should be given to using ANC as the most appropriate ecological indicator for this purpose, in that it provides the most stable metric that is highly associated with the water quality properties that are directly responsible for the principal

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adverse effects associated with aquatic acidification: fish mortality and reduced aquatic species diversity.

The PA developed such a form, using a simple equation to calculate an AAI value in terms of the ambient air indicators of oxides and nitrogen and sulfur and the relevant ecological and atmospheric factors that modify the relationships between the ambient air indicators and ANC. Recognizing the spatial variability of such factors across the U.S., the PA concludes it is appropriate to divide the country into ecologically relevant regions, characterized as acid-sensitive or relatively non-acid-sensitive, and specify the value of each of the factors in the AAI equation for each such region. Omernik ecoregions, level III, are identified as the appropriate set of regions over which to define the AAI. There are 84 such ecoregions that cover the continental U.S. This set of ecoregions is based on grouping a variety of vegetation, geological, and hydrological attributes that are directly relevant to aquatic acidification assessments and that allow for a practical application of an aquatic acidification standard on a national scale.

The PA defines AAI by the following equation: $AAI = F1 - F2 - F3[NO_y] - F4[SO_x]$. Factors F1 through F4 would be defined for each ecoregion by specifying ecoregion-specific values for each factor based on monitored or modeled data that are representative of each ecoregion. The F1 factor is also defined by a target ANC value. More specifically:

(1) F1 reflects a relative measure of an ecosystem's ability to neutralize acidifying deposition. The value of F1 for each ecoregion would be based on a representative critical load for the ecoregion associated with a single national target ANC level, as well as on a representative runoff rate. The representative runoff rate, which is also used in specifying values for the other factors, would be the median value of the distributions of runoff rates within the ecoregion. The representative critical load would be derived from

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a distribution of critical loads calculated for each water body in the ecoregion for which sufficient water quality and hydrology data are available. The representative critical load would be defined by selecting a specific percentile of the distribution.

In identifying a range of percentiles that are appropriate to consider for this purpose, regions categorized as acid sensitive were considered separately from regions categorized as relatively non-acid sensitive. For acid sensitive regions, the PA concludes that consideration should be given to selecting a percentile from within the range of the 70th to the 90th percentile. The lower end of this range was selected to be appreciably above the median value so as to ensure that the critical load would be representative of the population of relatively more acid sensitive water bodies within the region, while the upper end was selected to avoid the use of a critical load from the extreme tail of the distribution which is subject to a high degree of variability and potential outliers. For relatively non-acid sensitive regions, the PA concludes that consideration should be given to selecting the 50th percentile to best represent the distribution of water bodies within such a region, or alternatively to using the median critical load of all relatively non-acid sensitive areas, recognizing that such areas are far less frequently evaluated than acid sensitive areas. Using either of these approaches would avoid characterizing a generally non-acid-sensitive region with a critical load that is representative of relatively acid sensitive water bodies that may exist within a generally non-acid sensitive region.

(2) F2 reflects the deposition of reduced nitrogen. Consideration should be given to specifying the value of F2 for each region based on the averaged modeled value across the region, using national CMAQ modeling that has been conducted by EPA. Consideration could also be given to alternative approaches to specifying this value, such

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as the use of more localized and/or contemporaneous modeling in areas where this term is likely to be particularly variable and important.

(3) F3 and F4 reflect transference ratios that convert ambient air concentrations of NO_y and SO_x, respectively, into related deposition of nitrogen and sulfur. Consideration should be given to specifying the values for F3 and F4 for each region based on CMAQ modeling results averaged across the region. We conclude that specifying the values or the transference ratios based on CMAQ modeling results alone is preferred to an alternative approach that combines CMAQ model estimates with observational data.

(4) The terms [NO_y] and [SO_x] reflect ambient air concentrations measured at monitoring sites within each region.

Using the equation, a value of AAI can be calculated for any measured values of ambient NO_y and SO_x. For such a NAAQS, the Administrator would set a single, national value for the level of the AAI used to determine achievement of the NAAQS, as discussed below in section III.D. The ecoregion-specific values for factors F1 through F4 would be specified by EPA based on the most recent data and CMAQ model simulations, and codified as part of such a standard. These factors would be reviewed and updated as appropriate in the context of each periodic review of the NAAQS.

The PA developed specific F factors for each ecoregion based on the approach discussed above, using alternative percentiles and alternative national target ANC levels. The results of this analysis for ecoregions characterized as acid sensitive are presented in Table 7-1a-d in the PA.

C. Averaging Time

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As discussed in section 7.3 of the PA, aquatic acidification can occur over both long- and short-term timescales. Long-term cumulative deposition of nitrogen and sulfur is reflected in the chronic acid-base balance of surface waters as indicated by measured annual ANC levels. Similarly, the use of steady state critical load modeling, which generates critical loads in terms of annual cumulative deposition of nitrogen and sulfur, means that the focus of ecological effects studies based on critical loads is on the long-term equilibrium status of water quality in aquatic ecosystems. Much of the evidence of adverse ecological effects associated with aquatic acidification, as discussed above in section II.A, is associated with chronically low ANC levels. Protection against a chronic ANC level that is too low is provided by reducing overall annual average deposition levels for nitrogen and sulfur.

Reflecting this focus on long-term acidifying deposition, the PA developed the AAI that links ambient air indicators to deposition-related ecological effects, in terms of several factors, F1 through F4. As discussed above, these factors are all calculated as annual average values, whether based on water quality and hydrology data or on CMAQ model simulations. In the context of a standard defined in terms of the AAI, the PA concludes that it is appropriate to consider the same annual averaging time for the ambient air indicators as is used for the factors in the AAI equation.

We also recognize that short-term (i.e., hours or days) episodic changes in water chemistry, often due to changes in the hydrologic flow paths, can have important biological effects in aquatic ecosystems. Such short-term changes in water chemistry are termed "episodic acidification." Some streams may have chronic or base flow chemistry that is generally healthy for aquatic biota, but may be subject to occasional acidic episodes with potentially lethal

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consequences. Thus, short-term episodic ecological effects can occur even in the absence of long-term chronic acidification effects.

Episodic declines in pH and ANC are nearly ubiquitous in drainage waters throughout the eastern U.S. Episodic acidification can result from several mechanisms related to changes in hydrologic flow paths. For example, snow can store nitrogen deposited throughout the winter and snowmelt can then release this stored nitrogen, together with nitrogen derived from nitrification in the soil itself, in a pulse that leads to episodic acidification in the absence of increased deposition during the actual episodic acidification event. The PA notes that inputs of nitrogen and sulfur from snowpack and atmospheric deposition largely cycle through soil. As a result, short-term direct deposition inputs are not necessarily important in episodic acidification. Thus, as noted in chapter 3 of the ISA, protection against episodic acidity events can be achieved by establishing a higher chronic ANC level.

Taken together, the above considerations support the conclusion that it is appropriate to consider the use of a long-term average for the ambient air indicators NO_y and SO_x for an aquatic acidification standard defined in terms of the AAI. The use of an annual averaging time for NO_y and SO_x concentrations would be appropriate to provide protection against low chronic ANC levels, which in turn would protect against both long-term acidification and acute acidic episodes.

The PA has also considered interannual variability in both ambient air quality and in precipitation, which is directly related to the deposition of oxides of nitrogen and sulfur from the ambient air. While ambient air concentrations show year-to-year variability, often the year-to-year variability in precipitation is considerably greater, given the highly stochastic nature of precipitation. The use of multiple years over which annual averages are determined would

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dampen the effects of interannual variability in both air quality and precipitation. For the ambient air indicators, the use of multiple-year averages would also add stability to calculations used to judge whether an area meets a standard defined in terms of the AAI. Consequently, the PA concludes that an annual averaging time based on the average of each year over a consecutive 3- to 5-year period is appropriate to consider for the ambient air indicators NO_y and SO_x. In reaching this conclusion, the PA notes that in its comments on the second draft PA, CASAC agreed that a 3- to 5-year averaging time was appropriate to consider (Russell and Samet, 2010b).

D. Level

As discussed above, the PA concludes that ANC is the ecological indicator best suited to reflect the sensitivity of aquatic ecosystems to acidifying deposition from oxides of nitrogen and sulfur in the ambient air. The ANC is an indicator of the aquatic acidification expected to occur given the natural buffering capacity of an ecosystem and the loadings of nitrogen and sulfur resulting from atmospheric deposition. Thus, the PA developed a new standard for aquatic acidification that is based on the use of chronic ANC as the ecological indicator as a component in the AAI.

The level of the standard would be defined in terms of a single, national value of the AAI. The standard would be met at a monitoring site when the multi-year average of the calculated annual values of the AAI was equal to or *above* the specified level of the standard.[13] The annual values of the AAI would be calculated based on the AAI equation using the assigned ecoregion-specific values for factors F1 through F4 and monitored annual average NO_y and SO_x concentrations. Since the AAI equation is based on chronic ANC as the ecological indicator, the

[13] Unlike other NAAQS, where the standard is met when the relevant value is at or below the level of the standard since a lower standard level is more protective, in this case a higher standard level is more protective.

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level chosen for the standard would reflect a target chronic ANC value. As noted above, the assigned F factors for each ecoregion would be determined by EPA in the rulemaking to set the NAAQS, based on water quality and hydrology data, CMAQ modeling, the selected percentile that is used to identify a representative critical load within the ecoregion, and the selected level of the standard. The combination of the form of the standard, discussed above in section III.B, defined by the AAI equation and the assigned values of the F factors in the equation, other elements of the standard including the ambient air indicators (section III.A) and their averaging time (section III.C), and the level of the standard determines the allowable levels of NO_y and SO_x in the ambient air within each ecoregion. All of the elements of the standard together determine the degree of protection from adverse aquatic acidification effects associated with oxides of nitrogen and sulfur in the ambient air. The level of the standard plays a central role in determining the degree of protection provided and is discussed below.

The PA focuses primarily on information that relates degrees of biological impairment associated with adverse ecological effects to aquatic ecosystems to alternative levels of ANC in reaching conclusions regarding the range of target ANC levels that is appropriate to consider for the level of the standard. The PA develops the rationale for identifying a range of target ANC levels that is appropriate to consider by addressing questions related to the following areas: (1) associations between ANC and pH levels to provide an initial bounding for the range of ANC values to be considered; (2) evidence that allows for the delineation of specific ANC ranges associated with varying degrees of severity of biological impairment ecological effects; (3) the role of ANC in affording protection against episodic acidity; (4) implications of the time lag response of ANC to changes in deposition; (5) past and current examples of target ANC values

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applied in environmental management practices; and (6) data linking public welfare benefits and ANC.

1. Association Between pH Levels and Target ANC Levels

As discussed above in section II.A and more fully in chapter 3 of the PA, specific levels of ANC are associated with differing levels of risk of biological impairment in aquatic ecosystems, with higher levels of ANC resulting in lower risk of ecosystem impacts, and lower ANC levels resulting in risk of both higher intensity of impacts and a broader set of impacts. While ANC is not the causal agent determining biological effects in aquatic ecosystem, it is a useful metric for determining the level at which a water body is protected against risks of acidification. There is a direct correlation between ANC and pH levels which, along with dissolved aluminum, are more closely linked to the biological causes of ecosystem response to acidification.

Because there is a direct correlation between ANC and pH levels, the selection of target ANC levels is informed in part through information on effects of pH as well as direct studies of effects related to ANC. Levels of pH are closely associated with ANC in the pH range of approximately 4.5 to 7. Within this range, higher ANC levels are associated with higher pH levels. At a pH level of approximately 4.5, further reductions in ANC generally do not correlate with pH, as pH levels remain at approximately 4.5 while ANC values fall substantially. Similarly, at a pH value of approximately 7, ANC values can continue to increase with no corresponding increase in pH. As pH is the primary causal indicator of effects related to aquatic acidification, this suggests that ANC values below approximately -50 μeq/L (the apparent point in the relationship between pH and ANC where pH reaches a minimum) are not likely to result in further damage. In addition, ANC values around and above approximately 100 μeq/L (the

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apparent region in the relationship where pH reaches a maximum) are not likely to confer additional protection. As a result, the initial focus in the PA was on target ANC values in the range of -50 to 100 μeq/L.

2. ANC Levels Related to Effects on Aquatic Ecosystems

As discussed above in section II.A, the number of fish species present in a water body has been shown to be positively correlated with the ANC level in the water, with higher values supporting a greater richness and diversity of fish species. The diversity and distribution of phyto-zooplankton communities also are positively correlated with ANC.

A summary of effects related to ANC ranges is shown above in Table II-1. Within the ANC range from approximately -50 to 100 μeq/L, linear and sigmoidal relationships are observed between ANC and ecosystem effects. On average, fish species richness is lower by one fish species for every 21 μeq/L decrease in ANC in Shenandoah National Park streams (ISA, section 3.2.3.4). As shown in Table II-1, ANC levels have been grouped into five categories related to expected ecological effects, including categories of acute concern (<0 μeq/L), severe concern (0-20 μeq/L), elevated concern (20-50 μeq/L), moderate concern (50-100 μeq/L), and low concern (>100 μeq/L). This categorization is supported by a large body of research completed throughout the eastern U.S. (Sullivan et al., 2006).

Water bodies with ANC values less than or equal to 0 μeq/L at based flow are chronically acidic. Such ANC levels can lead to complete loss of species and major changes in the ability of water bodies to support diverse biota, especially in water bodies that are highly sensitive to episodic acidification. Based on the above considerations, the PA has focused on target ANC levels no lower than 0 μeq/L.

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As discussed in the PA, biota generally are not harmed when ANC values are >100 μeq/L, due to the low probability that pH levels will be below 7. In the Adirondacks, the number of fish species also peaks at ANC values >100 μeq/L. This suggests that at ANC levels greater than 100 μeq/L, little risk from acidification exists in many aquatic ecosystems. At ANC levels below 100 μeq/L, overall health of aquatic communities can be maintained, although fish fitness and community diversity begin to decline. At ANC levels ranging from 100 down to 50 μeq/L, there is increasing likelihood that the fitness of sensitive species (e.g., brook trout, zooplankton) will begin to decline. When ANC concentrations are below 50 μeq/L, the probability of acidification increases substantially, and negative effects on aquatic biota are observed, including large reductions in diversity of fish species and changes in the health of fish populations, affecting reproductive ability and fitness, especially in water bodies that are affected by episodic acidification. While there is evidence that ANC levels above 50 can confer additional protection from adverse ecological effects associated with aquatic acidification in some sensitive ecosystems, the expectation that such incremental protection from adverse effects will continue up to an ANC level of 100 is substantially reduced. The PA concludes that the above considerations support a focus on target ANC levels up to a level greater than 50 μeq/L but below 100 μeq/L, such as up to a level of 75 μeq/L.

In considering the available scientific evidence, as summarized here and discussed in more detail in the ISA and REA, in its review of the second draft PA, CASAC expressed the following views about the range of biological responses that corresponds to this range of ANC levels (i.e., 0-100 μeq/L):

> There will likely be biological effects of acidification at higher ANC values within this range, and there are relatively insensitive organisms that are not impacted at ANC values at the low end of this range. Adverse effects of

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> acidification on aquatic biota are fairly certain at the low end of this range of ANC and incremental benefits of shifting waters to higher ANC become more uncertain at higher ANC levels. There is substantial confidence that there are adverse effects at ANC levels below 20 μeq/L, and reasonable confidence that there are adverse effects below 50 μeq/L. Levels of 50 μeq/L and higher would provide additional protection, but the Panel has less confidence in the significance of the incremental benefits as the level increases above 50 μeq/L. (Russell and Samet, 2010b)

The PA concludes that the above considerations, including the views of CASAC, provide support for focusing on target ANC levels in the range of 20 to 75 μeq/L.

3. Consideration of Episodic Acidity

As discussed in the PA, across the broad range of ANC values from 0 to 100 μeq/L, ANC affords protection against the likelihood of decreased pH (and associated increases in Al) during long or short periods. In general, the higher the ANC within this range, the lower the probability of reaching low pH levels where direct effects such as increased fish mortality occur, as shown in Table 3-1 of the PA. Accordingly, greater protection would be achieved by target chronic ANC values set high enough to avoid pH depression to levels associated with elevated risk.

The specific relationship between ANC and the probability of reaching pH levels of elevated risk varies by water body and fish species. The ANC levels below 20 μeq/L are generally associated with high probability of low pH, leading to death or loss of fitness of biota that are sensitive to acidification (US EPA, 2008, section 5.2.2.1; US EPA, 2009, section 5.2.1.2). At these levels, during episodes of high acidifying deposition, brook trout populations may experience lethal effects. In addition, the diversity and distribution of zooplankton communities decline sharply at ANC levels below 20 μeq/L. Overall, there is little uncertainty that significant effects on aquatic biota are occurring at ANC levels below 20 μeq/L.

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It is clear that at ANC levels approaching 0 μeq/L (Table II-1), there is significant impairment of sensitive aquatic ecosystems with almost complete loss of fish species. Avoiding ANC levels approaching 0 μeq/L is particularly relevant to episodic spikes in acidity that occur during periods of rapid snow melt and during and after major precipitation events. Since the ANC range considered in the PA reflects average, long-term base flow values, it is appropriate to consider protecting against episodic drops in ANC values to a level as low as 0 μeq/L. Staddard et al. (2003) noted on average a 30 μeq/L depression of ANC between spring and summer time values, indicating the need to maintain higher base flow ANC levels to protect against ANC levels below 0 μeq/L. The above considerations do not provide support for a target chronic ANC level as low as 0 μeq/L for a standard that would protect against significant harm to aquatic ecosystems, including harm from episodic acidification. The PA concludes that these considerations also support a lower end of the range for consideration no lower than 20 μeq/L**.**

The CASAC agreed with this conclusion in its comments on the second draft PA (Russell and Samet, 2010b). The CASAC noted that "there are clear and marked biological effects at ANC values near 0 μeq/L, so this is probably not an appropriate target value" for the AAI. With regard to the likelihood of impairment of aquatic ecosystems due to episodic acidification, in terms of specific target levels for chronic ANC, CASAC expressed the following view:

> Based on surface waters studied in the Northeast, decreases in ANC associated with snowmelt [are] approximately 50 μeq/L. Thus, based on these studies, a long term ANC target level of 75 μeq/L would generally guard against effects from episodic acidification down to a level of about 25 μeq/L. (Russell and Samet, 2010b)

4. Consideration of Ecosystem Response Time

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The PA notes that when considering a standard level to protect against aquatic acidification, it is appropriate to take into account both the time period to recovery as well as the potential for recovery in acid-sensitive ecoregions. Ecosystems become adversely impacted by acidifying deposition over long periods of time and have variable time frames and abilities to recover from such perturbations. Modeling presented in the REA (US EPA, 2009, section 4.2.4) shows the estimated ANC values for Adirondack lakes and Shenandoah streams under pre-acidification conditions and indicates that for a small percentage of lakes and streams, natural ANC levels would have been below 50 μeq/L. Therefore, for these water bodies, reductions in acidifying deposition are not likely to achieve an ANC of 50 μeq/L or greater. Conversely, for some lakes and streams the level of perturbation from long periods of acidifying deposition has resulted in very low ANC values compared to estimated natural conditions. For such water bodies, the time to recovery would be largely dependent on future inputs of acidifying deposition.

Setting a standard level in terms of a target chronic ANC level is based on the long-term response of aquatic ecosystems. The time required for a water body to achieve the target ANC level -- given a decrease in ambient air concentrations of NO_y and SO_x and related acidifying deposition such that the critical load for a target ANC is not exceeded – is often decades if not centuries. In recognition of the potential public welfare benefits of achieving the target ANC in a shorter time frame, the concept of target loads had been developed. Target loads represent the depositional loading that is expected to achieve a particular level of the ecological indicator by a given time. For example, to achieve an ANC level of 20 μeq/L by 2030, it might be necessary to specify a higher target ANC level of, for example, 50 μeq/L, such that the depositional loading would be reduced more quickly than would occur if the depositional loading was based on

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achieving a target ANC level of 20 μeq/L as a long-term equilibrium level. In this example, the target ANC of 50 μeq/L would ultimately be realized many years later.

The above considerations have implications for selecting an appropriate standard level, in that the standard level affects not only the ultimate degree of protection that would be afforded by the standard, but also the time frame in which such protection would be realized. However, the PA recognizes that there is a great deal of heterogeneity in response times among water bodies and that there is only very limited information from dynamic modeling that would help to quantify recovery time frames in areas across the country. As a consequence, quantification of a general relationship between critical loads associated with a specific long-term target ANC level and target loads associated with achieving the target ANC level within a specific time frame is not currently possible. Thus, while the time frame for recovery is an important consideration in selecting an appropriate range of levels to consider, the PA concludes that it can only be considered in a qualitative sense at this time.

5. Prior Examples of Target ANC Levels

A number of regional organizations, states, and international organizations have developed critical load frameworks to protect against acidification of sensitive aquatic ecosystems. In considering the appropriate range of target ANC levels for consideration in this review, it is informative to evaluate the target ANC levels selected by these different organizations, as well as the rationale provided in support of the selected levels. Chapter 4 of the PA provides a detailed discussion of how critical loads have been developed and used in other contexts. Specific target values and their rationales are summarized below.

The UNECE has developed critical loads in support of international emissions reduction agreements. As noted in chapter 4 of the PA, critical loads were established to protect 95 percent

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of surface waters in Europe from an ANC less than 20 μeq/L based on protection of brown trout. Individual countries have set alternative ANC targets; for example, Norway targets an ANC of 30 μeq/L based on protection of Atlantic salmon. Several states have established target ANC or pH values related to protection of lakes and streams from acidification. While recognizing that some lakes in the Adirondacks will have a naturally low pH, the state of New York has established a target pH value of 6.5 for lakes that are not naturally below 6.5. As noted above, this level is associated with an ANC value that is likely to be between 20 and 50 μeq/L or possibly higher. New Hampshire and Vermont have set ANC targets of 60 μeq/L and 50 μeq/L, respectively. Tennessee has established site-specific target ANC values based on assessments of natural acidity, with a default value of 50 μeq/L when specific data are not available.

Taken together, these policy responses to concerns about ecological effects associated with aquatic acidification indicate that target ANC values between 20 and 60 μeq/L have been selected by states and other nations to provide protection of lakes and streams in some of the more sensitive aquatic ecosystems.

6. Consideration of Public Welfare Benefits

The point at which effects on public welfare become adverse is not defined in the CAA. Characterizing a known or anticipated adverse effect to public welfare is an important component of developing any secondary NAAQS. According to the CAA, welfare effects include:

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

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Consideration of adversity to public welfare in the context of the secondary NAAQS for oxides of nitrogen and sulfur can be informed by information about losses in ecosystem services associated with acidifying deposition and the potential economic value of those losses, as summarized above in section II.C and discussed more fully in chapter 4 of the PA.

Ecosystem service losses at alternative ANC levels are difficult to enumerate. However, in general there are categories of ecosystem services, discussed in chapter 4 of the PA, that are related to the specific ecosystem damages expected to occur at alternative ANC levels. Losses in fish populations due to very low ANC (below 20 μeq/L) are likely associated with significant losses in value for recreational and subsistence fishers. Many acid sensitive lakes are located in areas with high levels of recreational fishing activity. For example, in the northeastern U.S., where nearly 8 percent of lakes are considered acidic, more than 9 percent of adults participate in freshwater fishing, with an estimated value of approximately $5 billion in 2006. This suggests that improvements in lake fish populations may be associated with significant recreational fishing value.

As discussed in the PA, inland surface waters also provide cultural services such as aesthetic and existence value and educational services. To the extent that piscivorous birds and other wildlife are harmed by the absence of fish in these waters, hunting and birdwatching activities are likely to be adversely affected. A case study of the value to New York residents of improving the health of lakes in the Adirondacks found significant willingness to pay for those improvements. When scaled to evaluate the improvement in lake health from achieving ANC values of either 20 or 50 μeq/L, the study implies benefits to the New York population roughly on the order of $300 - 900 million per year (in constant 2007$). The survey administered in this study recognized that participants were thinking about the full range of services provided by the

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lakes in question – not just the recreational fishing services. Therefore the estimates of willingness to pay include resident's benefits for potential hunting and birdwatching activities and other ancillary services. These results are just for New York populations. The PA concludes that if similar benefits exist for improvements in other acid sensitive lakes, the economic value to U.S. populations could be very substantial, suggesting that, at least by one measure of impact on public welfare, impacts associated with ANC less than 50 μeq/L may be adverse to public welfare.

7. Summary of Alternative Levels

Based on all the above considerations, the PA concludes that consideration should be given to a range of standard levels from 20 to 75 μeq/L. The available evidence indicates that target ANC levels below 20 μeq/L would be inadequate to protect against substantial ecological effects and potential catastrophic loss of ecosystem function in some sensitive aquatic ecosystems. While ecological effects occur at ANC levels below 50 μeq/L in some sensitive ecosystems, the degree and nature of those effects are less significant than at levels below 20 μeq/L. Levels at and above 50 μeq/L would be expected to provide additional protection, although uncertainties regarding the potential for additional protection from adverse ecological effects are much larger for target ANC levels above about 75 μeq/L, as effects are generally appreciably less sensitive to changes in ANC at such higher levels.

In reaching this conclusion in the PA, consideration was given to the extent to which a target ANC level within this range would protect against episodic as well as long-term ecological effects. Levels in the mid- to upper part of this range would be expected to provide greater protection against short-term, episodic peaks in aquatic acidification, while lower levels within this range would give more weight to protection from long-term rather than episodic

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acidification. Similarly, levels in the mid- to upper part of this range would be expected to result in shorter time periods for recovery given the lag in ecosystem response in some sensitive ecosystems relative to levels in the lower part of this range. The PA also notes that this range encompasses target ANC values that have been established by various States and regional and international organizations to protect against acidification of aquatic ecosystems.

The PA recognizes that the level of the standard together with the other elements of the standard, including the ambient air indicators, averaging time, and form, determine the overall protectiveness of the standard. Thus, consideration of a standard level should reflect the strengths and limitations of the evidence and assessments as well as the inherent uncertainties in the development of each of the elements of the standard. The implications of considering alternative standards, defined in terms of alternative combinations of levels and percentile values that are a critical component of factor F1 in the form of the standard, are discussed below in section III.E. Key uncertainties in the various components of the standard are summarized and considered below in section III.F.

E. Combined Alternative Levels and Forms

To provide some perspective on the implications of various alternative multi-pollutant, AAI-based standards, the PA presented the number of acid-sensitive ecoregions that would likely not meet various sets of alternative standards. The alternative standards considered were based on combinations of alternative target ANC levels, within the range of 20 to 75 μeq/L, and alternative forms, characterized by alternative representative percentiles within the range of the 70th to 90th percentile. These alternative standards are also defined in terms of the other elements of the standard: ambient air indicators NOy and SOx, discussed above in section III.A; other elements of the form of the standard, including ecoregion-specific values for factors F1 through

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F4 in the AAI equation, discussed above in section III.B.5; and an annual averaging time for NO_y and SO_x, discussed above in section III.C. With regard to the averaging time, the assessment did not consider multi-year averaging of the calculated annual AAI values due to data limitations, including, for example, the lack of CMAQ modeling for multiple consecutive years. In this assessment, we characterize an ecoregion as likely not meeting a given alternative standard if the calculated AAI value is less than the target ANC level of the standard, recognizing that higher AAI values are more protective than lower values.

The results of this assessment are presented in Table 7-1a-d in the PA for a subset of ecoregions including those characterized as acid sensitive. Calculated annual AAI values at the ecoregion level are shown for each alternative standard considered. Based on these AAI values, Table 7-2 in the PA summarizes the number of acid-sensitive ecoregions that would likely not meet each of the alternative standards considered.[14] Calculated AAI values for all ecoregions categorized as relatively non-acid sensitive are shown in Table D-5 in Appendix D of the PA. In all cases, these relatively non-acid sensitive ecoregions were estimated to meet all of the alternative standards considered in this assessment.

As described above, the AAI values presented in Table 7-1a-d of the PA are based in part on data from 2005 CMAQ model simulations, which was used to generate values for F2 through F4 in the AAI equation, as well as to estimate annual average ambient air concentrations of NO_y and SO_x that reflect recent air quality in the absence of currently available monitored concentrations in sensitive ecoregions across the country. Water quality and hydrology data from water bodies within each ecoregion were also used in calculating the AAI values. Such

[14] Tables 7-1a-d and 7-2 in the PA present assessment results for 29 ecoregions that had been initially characterized as acid sensitive. Subsequently, based on a broader set of criteria used to characterize ecoregions as acid sensitive, as discussed above in section III.B.5.a, the set of ecoregions characterized as acid sensitive was narrowed to include 22 ecoregions.

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data were initially used to calculate critical loads for each water body with sufficient data within an ecoregion so as to identify the nth percentile critical load representative of the ecoregion used in calculating the F1 factor for the ecoregion. As expected, the number of ecoregions that likely would not meet alternative standards increases with increasing percentile values and target ANC levels (US EPA, 2011, Table 7-2). Out of 22 acid-sensitive ecoregions, the number of ecoregions that would likely not meet the alternative standards ranges from 22 for the most protective alternative standard considered (75 μeq/L, 90th percentile) to 4 for the least protective alternative standard (20 μeq/L, 70th percentile). It is apparent that both the percentile and the level chosen have a strong influence, over the ranges considered, in determining the number of areas that would likely not meet this set of alternative standards.

The PA observes that there is one grouping of these acid-sensitive ecoregions that would likely not meet almost all combinations of level and form under consideration (US EPA, 2011, Table 7-2 and Appendix D). This group is made up of southern Appalachian mountain areas, including North Central Appalachians, 5.3.3; Ridge and Valley, 8.4.1; Central Appalachians, 8.4.2; Blue Ridge, 8.4.4; and Southwestern Appalachians, 8.4.9. In addition, these ecoregions exhibit the highest amounts of exceedance relative to alternative standards.

The Northern Appalachian and Atlantic Maritime Highlands (5.3.1), which includes the Adirondacks, and the Northern Lakes and Forests (5.2.1) of the upper midwest exhibit similar patterns with respect to in the role of level and percentile in identifying regions not likely to meet alternative standards, although there are considerably fewer cases compared to the regions in the Appalachians.

In the mountainous west, the Sierra Nevada (6.2.12), Idaho Batholith (6.2.15) and the Cascades (6.2.7) ecoregions likely would not meet alternative standards in fewer cases relative to

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eastern regions, with the Sierra Nevada ecoregion exhibiting relatively greater sensitivity compared to all western regions. Only in the upper part of the ranges of level and percentile do regions in the northern and central Rockies likely not meet alternative standards.

In considering these findings, the PA observes that the standard as defined by the AAI behaves in an intuitively logical manner. That is, an increase in ecoregions likely not to meet the standard is associated with higher alternative levels and percentiles, both of which contribute to a lower regionally representative critical load. Moreover, the areas of known adverse aquatic acidification effects are identified, mostly in high elevation regions or in the northern latitudes -- the Adirondacks, Shenandoahs, northern midwest lakes and the mountainous west. These results reflect the first application of a nationwide model that integrates water quality and atmospheric processes at a national scale and provides findings that are consistent with our basic understanding of the extent of aquatic acidification across the U.S. What is particularly noteworthy is that this model is not initialized with a starting ANC based on water quality data, which likely would result in a reproduction of water quality observations. Rather, this standard reflects the potential of the changes in atmospheric concentrations of NO_y and SO_x to induce long-term sustained changes in surface water systems. The PA notes that the fact that the patterns of adversity based on applying this standard are commensurate with what is observed in surface water systems provides confidence in the basic underlying formulation of the standard.

The PA notes that the Appalachian mountain regions merit further inspection as they stand out as areas with the largest relative exceedances from a national perspective. Water quality data from these regions as well as an emissions sensitivity CMAQ simulation were considered to better understand the simulated behavior of these regions. The maps and tables in appendix D of the PA include paired comparisons of the CMAQ 2005 and emissions sensitivity

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simulations. The emissions sensitivity simulation reflects domain-wide reductions in NO_y and SO_x emissions of 48 percent and 42 percent, respectively, relative to 2005 base year emissions. The PA assumes that this emissions sensitivity simulation is indicative of future conditions.

The emissions sensitivity results project that many of the regions that likely would not meet the alternative standards based on recent air quality, especially at alternative levels of 20 and 35 μeq/L, would likely meet such standards in the future year scenario for the Appalachian mountain regions. It is apparent that the AAI calculations are especially sensitive to changes in SOx emissions as the Appalachian regions have the highest SO_x concentrations and deposition rates (US EPA, 2011,section 2), and the AAI equation responds as expected to modeled reductions in SO_x. The emissions sensitivity scenario is a prospective application of the standard, in the sense that rules derived from the air quality management process result in reductions of NO_y and SO_x emissions. Expected emission changes over the next two decades should be far greater than the 42 percent and 48 percent, respectively, SO_x and NO_y reductions used in this analysis, with a consequent further reduction in areas that would likely not meet alternative standards.

The Appalachian mountain regions generally have low DOC levels, average runoff rates, moderately low base cation supply and highly elevated sulfate concentrations. Collectively, those attributes do not suggest naturally acidic conditions as the availability of anthropogenic contributions of mineral acids is likely responsible for observed low ANC values in those regions.

The PA notes the Sierra Nevada region as an interesting case study, as it has some of the lowest critical load values nationally (US EPA, 2011, Table D-3). Water quality data indicate extremely low sulfate, as expected given the relatively low SO_2 emissions in the western U.S.

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Extremely low base cation supply and low Neco, which mitigate the effect of nitrogen deposition, explain the low critical load values. Low Neco values appear to associate well with high elevation western U.S. regions, perhaps reflecting the more arid and reduced vegetation density relative to eastern U.S. regions. The proximity to high level nitrogen emissions combined with very low base cation supply explains the cases where the Sierra region likely does not meet alternative standards. Because Neco values are low in the Sierras, the system responds effectively to reductions of NO_x emissions, as illustrated in the maps and tables of Appendix D of the PA. Although Neco affords protection from the acidifying effects of nitrogen deposition, the availability of excessive nitrogen neutralization capacity also means that reductions in nitrogen are not as effective as reductions in SO_x in reducing the calculated AAI.

In reviewing these results, the PA observes that the analysis of the alternative combinations of level and form presented provide context for considering the impact of different standards. Since the AAI equation has been newly developed in the PA, these examples of estimated exceedances help to address the question of whether the AAI equation responds in a reasonable manner with regard to identifying areas of concern and to prospective changes in atmospheric conditions likely to result from future emissions reduction strategies. The PA concludes that the behavior of the AAI calculations is both reasonable and explainable, which the PA concludes serves to increase confidence in considering a standard defined in terms of the AAI.

F. Characterization of Uncertainties

This section summarizes discussions of the results of analyses and assessments, presented more fully in the PA (US EPA, 2011, section 7.6 and Appendices F and G), intended to address the relative confidence associated with the linked atmospheric-ecological effects system

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described above. An overview of uncertainties is presented in the context of the major structural components underlying the standard, as well as with regard to areas of relatively high uncertainty. The section closes with a discussion of data gaps and uncertainties associated with the use of ecological and atmospheric modeling to specify the factors in the AAI equation, which can be used to guide future field programs and longer-term research efforts.

1. Overview of Uncertainty

As discussed in the PA (US EPA, 2011,Table 7-3), there is relatively low uncertainty with regard to the conceptual formulation of the overall structure of the AAI-based standard that incorporates the major associations linking biological effects to air concentrations. Based on the strength of the evidence that links species richness and mortality to water quality, the associations are strongly causal and without any obvious confounding influence. The strong association between the ecosystem indicator (ANC) and the causative water chemistry species (dissolved aluminum and hydrogen ion) reinforces the confidence in the linkage between deposition of nitrogen and sulfur and effects. This strong association between ANC and effects is supported by a sound mechanistic foundation between deposition and ANC. The same mechanistic strength holds true for the relationship between ambient air levels of nitrogen and sulfur and deposition, which completes the linkage from ambient air indicators through deposition to ecological effects.

There are relatively higher uncertainties, however, in considering specific elements within the structure of an AAI-based standard, including the deposition of SO_x, NO_y, and NH_x as well as the critical load-related component, each of which can vary within and across ecoregions. Overall system uncertainty relates not just to the uncertainty in each such element, but also to the combined uncertainties that result from linking these elements together within the AAI-based

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structure. Some of these elements – including, for example, dry deposition, pre-industrial base cation production, and reduced nitrogen deposition – are estimated with less confidence than other elements (US EPA, 2011, Table 7.3). The uncertainties associated with all of these elements, and the combination of these elements through the AAI equation, are discussed below and in the following sections related to measured data gaps and modeled processes for both air quality and water quality.

The lack of observed dry deposition data is constrained by resources and the lack of efficient measurement technologies. Progress in reducing uncertainties in dry deposition will depend on improved atmospheric concentration data and direct deposition flux measurements of the relevant suite of NO_y and SO_x species.

Pre-industrial base cation productivity by definition is not observable. Contemporary observations and inter-model comparisons are useful tools that would help reduce the uncertainty in estimates of preindustrial base cation productivity used in the AAI equation. In characterizing contemporary base cation flux using basic water quality measurements (i.e., major anion and cation species as defined in equation 2.11 in the PA), it is reasonable to assume that a major component of contemporary base cation flux is associated with pre-industrial weathering rates. To the extent that multiple models converge on similar solutions, greater confidence in estimating pre-industrial base cation production would be achieved.

Characterization of NH_x deposition has been evolving over the last decade. The relatively high uncertainty in characterizing NH_x deposition is due to both the lack of field measurements and the inherent complexity of characterizing NH_x with respect to source emissions and dry deposition. Because ammonia emissions are generated through a combination of man-made and biological activities, and ammonia is semi-volatile, the ability to characterize

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spatial and temporal distributions of NH_x concentrations and deposition patterns is challenging. While direct measurement of NH_x deposition is resource intensive because of the diffuse nature of sources (i.e., area-wide and non-point sources), there have been more frequent deposition flux studies, relative to other nitrogen species, that enable the estimation of both emissions and dry deposition. Also, while ammonia has a relatively high deposition velocity and traditionally was thought to deposit close to the emissions release areas, the semi-volatile nature of ammonia results in re-entrainment back into the lower boundary layer resulting in a more dispersed concentration pattern exhibiting transport type characteristics similar to longer lived atmospheric species. These inherent complexities in source characterization and ambient concentration patterns raise the uncertainty level of NH_x in general. However, the PA notes that progress is being made in measuring ammonia with cost efficient samplers and anticipates the gradual evolution of a spatially robust ammonia sampling network that would help support analyses to reduce underlying uncertainties in NH_x deposition. Also, from an aquatic acidification perspective, NH_x is not as important a driver as NO_y and SO_x in the mountainous areas in the eastern U.S. However, the relative importance of NH_x is likely to increase over time, in light of air quality rules in place designed to reduce emissions of NO_y and SO_x.

2. Uncertainties Associated with Data Gaps

In summarizing uncertainties with respect to available measurement data and the use of ecological and atmospheric models, the PA indentified data gaps and model uncertainties in relative terms by comparing, for example, the relative richness of data between geographic areas or environmental media. With regard to relevant air quality measurements, the PA notes that such measurements are relatively sparse in the western U.S. While the spatial extent of CASTNET coverage has gradually incorporated western U.S. locations with support from the

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NPS, the relative density of monitoring sites is much less than that in the eastern U.S. This relative disparity in spatial density of monitors is exacerbated as air quality patterns in the mountainous west generally exhibit greater spatial heterogeneity due to dramatic elevation gradients that impact meteorology and air mass flow patterns. Similarly, water quality data coverage is far more comprehensive in the eastern U.S. relative to the west

Measurements of NO_y notably are lacking in both eastern and western acid-sensitive ecoregions. This adds uncertainty to the use of the AAI equation as the lack of NO_y data limits efforts to evaluate air quality modeling of NO_y that is the basis for quantifying factor F3 in the AAI equation. The lack of NO_y measurements also limits efforts to characterize the variability and representativeness of modeled NO_y concentrations within and across ecoregions. Currently, the Agency's ability to define the protection likely to be afforded by alternative standards (in terms of alternative levels and percentiles) is compromised by the lack of a full set of ambient air quality indicator measurements, notably including NO_y, throughout sensitive ecoregions across the U.S.

Further, obtaining measurement of the dominant species that comprise NO_y (HNO_3, true NO_2, NO, p–NO_3, and PAN) would be useful to evaluate performance of NO_y samplers. Beyond the more well known dominant components of NO_y, research efforts would be needed to characterize total reactive nitrogen that may include significant amounts of organically-bound nitrogen (beyond PAN) which is poorly understood with regard to emission sources and concentration levels.

Field measurements of NH_x have been extremely limited, but have begun to be enhanced through the NADP's passive ammonia network (AMoN). The AMoN measures ammonia at over 50 sites, with more than 35 at CASTNET locations. Enhanced spatial coverage of reduced

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nitrogen measurements, particularly to understand within and across ecoregion variability, and the inclusion of some continuous observations would provide a better understanding of the uncertainty in the F2 factor in the AAI equation and of the representativeness of modeled NH_x deposition within and across ecoregions.

With regard to water quality data, the PA notes that such data are typically limited relative to air quality data sets, and are also relatively sparse in the western U.S. The TIME/LTM water quality sampling program in the eastern U.S. (as described in chapter 2 of the PA) is an appropriate complement to national air monitoring programs as it affords consistency across water bodies in terms of sampling frequency and analysis protocols. Consideration should be given to extending the TIME/LTM design to all acid sensitive ecoregions, with priority for areas in the western mountains that are data limited and showing initial signs of adversity particularly with respect to aquatic acidification. The lack of a regulatory requirement for TIME/LTM often jeopardizes funding support of this resource that is especially valuable and cost effective. While there are several state and local agency water quality data bases, it is unclear the extent to which differences in sampling, chemical analysis and reporting protocols would impact the use of such data for the purpose of better understanding the degree of protectiveness that would be afforded by an AAI-based standard within sensitive ecoregions across the country. In addition, our understanding of water quality in Alaska and Hawaii and the acid sensitivity of their ecoregions is particularly limited.

Water quality data and modeling support the standard setting process. As more water bodies are sampled, the critical load data bases would expand, enabling clearer delineation of ecoregion representative critical loads in terms of the nth percentile. This would provide more refined characterization of the degree of protection afforded by a given standard. Longer term,

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the availability of water quality trend data (annual to monthly sampled) would support accountability assessments that examine if an ecoregion's response to air management efforts is as predicted by earlier model forecasting. The most obvious example is the long-term response of water quality ANC change to changes in calculated AAI, deposition, ambient NO_y and SO_x concentrations, and emissions. In addition, water quality trends data provide a basis for evaluating and improving the parameterizations of processes in critical load models applied at the ecoregion scale related to nitrogen retention and base cation supply. A better understanding of soil processes, especially in the southern Appalachians, would enhance efforts to examine the variability within ecoregions of the soil-based adsorption and exchange processes which moderate the supply of major cations and anions to surface waters and strongly influence the response of surface water ANC to changes in deposition of nitrogen and sulfur.

3. Uncertainties in Modeled Processes

As discussed in the PA, from an uncertainty perspective, gaps in field measurement data are related to uncertainties in modeled processes and in the specific application of such models. As noted above, processes that are embodied in an AAI-based standard are modeled using the CMAQ atmospheric model and steady state ecological models. These models are characterized in the ISA as being well established and they have undergone extensive peer review. Nonetheless, the application of these models for purposes of specifying the factors in the AAI equation, on an ecoregion scale, is a new application that introduces uncertainties, as noted below, especially in areas with limited observational data that can be used to evaluate this specific application. Understanding uncertainties in relevant modeled process thus involves consideration of the uncertainties associated with applying each model as well as the

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combination of these uncertainties as the models are applied in combination within the AAI framework.

With regard to the application of CMAQ for purposes of use in an AAI-based standard, the modeling of dry deposition has been identified as having a relatively high degree of uncertainty. Due to a combination of system complexity and resource constraints, there is no routine observational basis for directly comparing modeled dry deposition and measurements. Periodic dry deposition flux experiments covering a variety of vegetation, surfaces and meteorology across seasons would enable a more robust evaluation of modeled deposition of nitrogen and sulfur. Given the difficulty in acquiring dry deposition observations, it becomes especially important to evaluate the model's ability to capture temporal and spatial ambient air patterns of individual nitrogen and sulfur species which are used to drive dry deposition calculations in models. For example, reducing a generally acknowledged positive bias in model-predicted SO_2 relative to observations is especially relevant to the AAI-based standard, as SO_2 deposition is a dominant contributor to total acidifying deposition in the eastern U.S. With respect to oxidized nitrogen, observations of individual NO_y species are important as air quality models calculate the individual deposition of each species. The modeled transference ratios, T_{NOy} and T_{SOx} used in factors F3 and F4 rely on CMAQ's ability to characterize both deposition and concentration. Consequently, a better understanding of the variability of these factors within and across ecoregions could be achieved by improved availability of measured ambient concentrations and deposition observations.

Steady state biogeochemical ecosystem modeling is used to develop critical load estimates that are incorporated in the AAI equation through factor F1. Consequently, the PA notes that an estimate of the temporal response of surface water ANC to deposition and air

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concentration changes is not directly available. Lacking a predicted temporal response impairs the ability to conduct accountability assessments down to the effects level. Accountability assessments would examine the response of each step in the emissions source through air concentration – deposition -- surface water quality – biota continuum. The steady state assumption at the ecosystem level does not impair accountability assessments through the air concentration/deposition range of that continuum. However, in using steady state ecosystem modeling, several assumptions are made relative to the long-term importance of processes related to soil adsorption of major ions and ecosystem nitrogen dynamics. Because these models often were developed and applied in glaciated areas with relatively thin and organically rich soils, their applicability is relatively more uncertain in areas such as those in the non-glaciated clay-based soil regions of the central Appalachians. Consequently, it is desirable to develop the information bases to drive simple dynamic ecosystem models that incorporate more detailed treatment of subsurface processes, such as adsorption and exchange processes and sulfate absorption.

4. Applying Knowledge of Uncertainties

An understanding of the relative uncertainties in a system assists in setting priorities for data collection efforts and research, with the expectation that such efforts would reduce uncertainties over time and afford greater confidence in applications of an AAI-based standard. Because of the uniquely wide breadth of pollutants and environmental media addressed by an AAI-based multi-pollutant standard, there are a wide range of uncertainties that are important to consider relative to single pollutant standards that typically address only direct effects of ambient air exposures. For an AAI-based standard, a reduction of the uncertainties across the various

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modeled processes at the ecoregion scale would lead to greater confidence in the degree of protection afforded by the standard.

The PA notes that there is generally low uncertainty with regard to the conceptual development and related major components of this standard. In recognizing the scientific soundness of the basic structure of this standard, the PA notes that future efforts would be appropriately directed at expanding the availability of relevant data for ecoregion-specific evaluation and application of the relevant modeling of ecological and atmospheric processes, as identified above. Such efforts would further support consideration of an AAI-based standard and would guide field studies and analyses designed to improve the longer-term confidence in such a standard.

G. CASAC Advice

The CASAC has advised EPA concerning the ISA, the REA, and the PA. The CASAC has endorsed EPA's interpretation of the science embodied in the ISA and the assessment approaches and conclusions incorporated in the REA.

Most recently, CASAC has considered the information in the final PA in providing its recommendations on the review of the new multi-pollutant standard developed in that document and discussed above (Russell and Samet, 2011a). In so doing, CASAC has expressed general support for the conceptual framework of the standard based on the underlying scientific information, as well as for the conclusions in the PA with regard to indicators, form, averaging time, and level of the standard that are appropriate for consideration by the Agency in reaching decisions on the review of the secondary NAAQS for oxides of nitrogen and sulfur:

> The final *Policy Assessment* clearly sets out the basis for the recommended ranges for each of the four elements (indicator, averaging time, level and form) of a potential NAAQS that uses ambient air indicators to address the combined effects

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> of oxides of nitrogen and oxides of sulfur on aquatic ecosystems, primarily streams and lakes. As requested in our previous letters, the *Policy Assessment* also describes the implications of choosing specific combinations of elements and provides numerous maps and tabular estimates of the spatial extent and degree of severity of NAAQS exceedances expected to result from possible combinations of the elements of the standard.
>
> We believe this final PA is appropriate for use in determining a secondary standard to help protect aquatic ecosystems from acidifying deposition of oxides of sulfur and nitrogen. EPA staff has done a commendable job developing the innovative Aquatic Acidification Index (AAI), which provides a framework for a national standard based on ambient concentrations that also takes into account regional differences in sensitivities of ecosystems across the country to effects of acidifying deposition. (Russell and Samet, 2011a)

The CASAC also recommended that as EPA moves forward in the regulatory process "some attention should be given to our residual concern that the available data may reflect the more sensitive water bodies and thus, the selection of percentiles of waterbodies to be protected could be conservatively biased" (Russell and Samet, 2011a). In addition, CASAC found some improvements could be made to the uncertainty analysis, as noted below. With respect to indicators, CASAC supports the use of SO_x and NO_y as ambient air indicators (discussed above in section III.A) and ANC as the ecological indicator (discussed above in section III.B.1):

The use of NO_y and SO_x as atmospheric indicators of oxides of nitrogen and sulfur atmospheric concentrations is well justified. The use in the AAI of NO_y and SO_x as atmospheric indicators of oxides of nitrogen and sulfur concentrations is useful and corresponds with other efforts by EPA. As we have stated previously, CASAC also agrees that ANC is the most appropriate ecological indicator of aquatic ecosystem response and resiliency to acidification (Russell and Samet, 2011a).

With respect to the form of the standard (discussed above in section III.B), CASAC stated the following:

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> EPA has developed the AAI, an innovative "form" of the NAAQS itself that incorporates the multi-pollutant, multi-media, environmentally modified, geographically variable nature of SOx/NOy deposition-related aquatic acidification effects. With the caveats noted below, CASAC believes that this form of the NAAQS as described in the final Policy Assessment is consistent with and directly reflective of current scientific understanding of effects of acidifying deposition on aquatic ecosystems. (Russell and Samet, 2011a)

> CASAC agrees that the spatial components of the form in the Policy Assessment are reasonable and that use of Omernick's ecoregions (Level III) is appropriate for a secondary NAAQs intended to protect the aquatic environment from acidification . . . (Russell and Samet, 2011a)

The "caveats" noted by CASAC include a recognition of the importance of continuing to evaluate the performance of the CMAQ and ecological models to account for model uncertainties and to make the model-dependent factors in the AAI more transparent. In addition, CASAC noted that the role of DOC and its effects on ANC would benefit from further refinement and clarification (Russell and Samet, 2011a). While CASAC expressed the view that the "division of ecoregions into 'sensitive' and 'non-sensitive' subsets, with a more protective percentile applied to the sensitive areas, also seems reasonable" (Russell and Samet, 2011a), CASAC also noted that there was the need for greater clarity in specifying how appropriate screening criteria would be applied in assigning ecoregions to these categories. Further, CASAC identified potential biases in critical load calculations and in the regional representativeness of available water chemistry data, leading to the observation that a given percentile of the distribution of estimated critical loads may be protective of a higher percentage of surface waters in some regions (Russell and Samet, 2011a).

With respect to averaging time (discussed above in section III.C), CASAC stated the following:

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> Considering the cumulative nature of the long-term adverse ecological effects and the year-to-year variability of atmospheric conditions (mainly in the amount of precipitation), CASAC concurs with EPA that an averaging time of three to five years for the AAI parameters is appropriate. A longer averaging time would mask possible trends of AAI, while a shorter averaging time would make the AAI being more influenced by the conditions of the particular years selected. (Russell and Samet, 2011a)

With respect to level as well as the combination of level and form as they are presented as alternative standards (discussed above in sections III.D-E), CASAC stated the following:

> CASAC agrees with EPA staff's recommendation that the "level" of the alternative AAI standards should be within the range of 20 and 75 μeq/L. We also recognize that both the "level" and the form of any AAI standard are so closely linked in their effectiveness that these two elements should be considered together. (Russell and Samet, 2011a)
>
> When considered in isolation, it is difficult to evaluate the logic or implications of selecting from percentiles (70th to 90th) of the distribution of estimated critical loads for lakes in sensitive ecoregions to determine an acceptable amount of deposition for a given ecoregion. However, when these percentile ranges are combined with alternative levels within the staff-recommended ANC range of 20 to 75 microequivalents per liter (μeq/L), the results using the AAI point to the ecoregions across the country that would be expected to require additional protection from acidifying deposition. Reasonable choices were made in developing the form. The number of acid sensitive regions not likely to meet the standard will be affected both by choice of ANC level and the percentile of the distribution of critical loads for lakes to meet alternative ANC levels in each region. These combined recommendations provide the Administrator with a broad but reasonable range of minimally to substantially protective options for the standard. (Russell and Samet, 2011a)

CASAC also commented on EPA's uncertainty analysis, and provided advice on areas requiring further clarification in the proposed rule and future research. The CASAC found it "difficult to judge the adequacy of the uncertainty analysis performed by EPA because of lack of details on data inputs and the methodology used, and lack of clarity in presentation" (Russell and Samet, 2011a). In particular, CASAC identified the need for more thorough model evaluations of critical load and atmospheric modeling, recognizing the important role of models as they are

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incorporated in the form of the standard. In light of the innovative nature of the standard developed in the PA, CASAC identified "a number of areas that should be the focus of further research" (Russell and Samet, 2011a). While CASAC recognized that EPA staff was able to address some of the issues in the PA, they also noted areas "that would benefit from further study or consideration in potential revisions or modifications to the form of the standard." Such research areas include "sulfur retention and mobilization in the soils, aluminum availability, soil versus water acidification and ecosystem recovery times." Further, CASAC encouraged future efforts to monitor individual ambient nitrogen species, which would help inform further CMAQ evaluations and the specification of model-derived elements in the AAI equation (Russell and Samet, 2011a).

H. Administrator's Proposed Conclusions

Having concluded that the existing NO_2 and SO_2 secondary standards are neither sufficiently protective nor appropriate to address deposition-related effects associated with oxides of nitrogen and sulfur (section II.D above), the Administrator has considered whether it is appropriate at this time to set a new multi-pollutant standard for that purpose, with a structure that would better reflect the available science regarding acid deposition. In considering this, she recognizes that such an appropriate standard, for purposes of section 109(b) and (d) of the CAA,[15], must in her judgment be requisite to protect public welfare, such that it would be neither more nor less stringent that necessary for that purpose. In particular, she has focused on the new standard developed in the PA and reviewed by CASAC, as discussed above. In so doing, the Administrator first considered the extent to which there is a scientific basis for development of

[15] Section 109(d)(1) requires that "…the Administrator shall complete a thorough review…and shall make such revisions in such criteria and standards and promulgate such new standards as may be *appropriate* under…subsection 109(b) of this section." [emphasis added]

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such a standard, specifically with regard to a standard that would provide protection from deposition-related aquatic acidification in sensitive aquatic ecosystems in areas across the country. As discussed above, the Administrator notes that the ISA concludes that the available scientific evidence is sufficient to infer a causal relationship between acidifying deposition of nitrogen and sulfur in aquatic ecosystems, and that the deposition of oxides of nitrogen and sulfur both cause such acidification under current conditions in the U.S. Further, the ISA concludes that there are well-established water quality and biological indicators of aquatic acidification as well as well-established models that address deposition, water quality, and effects on ecosystem biota, and that ecosystem sensitivity to acidification varies across the country according to present and historic nitrogen and sulfur deposition as well as geologic, soil, vegetative, and hydrologic factors. Based on these considerations, the Administrator agrees with the conclusion in the PA, and supported by CASAC, that there is a strong scientific basis for development of a standard with the general structure presented in the PA.

The Administrator also recognizes that the conceptual framework for an ecologically relevant, multi-pollutant standard, which was initially explored in the REA and further developed in the PA, builds on the information in the ISA. She notes that the structure of the standard addresses the combined effects of deposition from oxides of nitrogen and sulfur by characterizing the linkages between ambient concentrations, deposition, and aquatic acidification, and that the structure of the standard takes into account relevant variations in these linkages across the country. She recognizes that while the standard is innovative and unique, the structure of the standard is well grounded in the science underlying the relationships between ambient concentrations of oxides of nitrogen and sulfur and the aquatic acidification related to deposition of nitrogen and sulfur associated with such ambient concentrations.

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While the Administrator recognizes the strong scientific foundation for the structure of an AAI-based standard, she also recognizes that the standard depends on atmospheric and ecological modeling, based on appropriate data, to specify the terms of an equation that incorporates the linkages between ambient concentrations, deposition, and aquatic acidification. This equation, which defines an aquatic acidification index (AAI), has the effect of translating spatially variable ecological effects into a potential national standard. With respect to establishing the specific terms of this equation, there are a number of inherent uncertainties and complexities that are relevant to the question of whether it is appropriate under section 109 to set a specific AAI-based standard at this time, recognizing that such a standard must be requisite to protect public welfare without being either more or less stringent than necessary for this purpose. As discussed above, these uncertainties and complexities generally relate not to the structure of the standard, but to the quantification of the various elements of the standard, such as the F factors discussed earlier in this section and their representativeness at an ecoregion scale. These uncertainties and complexities currently limit efforts to characterize the degree of protectiveness that would be afforded by such a standard, within the ranges of levels and forms identified in the PA, and the representativeness of F factors in the AAI equation described above and in the PA. These important uncertainties have been generally categorized as limitations in available field data as well as uncertainties that are related to reliance on the application of ecological and atmospheric modeling at the ecoregion scale to specify the various elements of the AAI.

With regard to data limitations, the Administrator observes that there are several important limitations in the available data upon which elements of the AAI are based. For example, while ambient measurements of NO_y are made as part of a national monitoring network, the monitors are not located in locations that are representative of sensitive aquatic

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ecosystems. While air and water quality data are generally available in areas in the eastern U.S., there is relatively sparse coverage in mountainous western areas where a number of sensitive aquatic ecosystems are located. Further, even in areas where relevant data are available, small sample sizes impede efforts to characterize the representativeness of the available data, which was noted by CASAC as being of particular concern. Also, measurements of reduced forms of nitrogen are available from only a small number of monitoring sites, and emission inventories for reduced forms of nitrogen used in atmospheric modeling are subject to considerable uncertainty.

With regard to uncertainties related to the use of ecological and atmospheric modeling, the Administrator notes in particular that model results are difficult to evaluate due to a lack of relevant observational data. For example, relatively large uncertainties are introduced by a lack of data with regard to pre-industrial environmental conditions and other parameters that are necessary inputs to critical load models that are the basis for factor F1 in the AAI equation. Also, observational data are not generally available to evaluate the modeled relationships between nitrogen and sulfur in the ambient air and associated deposition, which are the basis for the other factors (i.e., F2, F3, and F4) in the AAI equation.

In combination, these limitations and uncertainties result in a considerable degree of uncertainty as to how well the quantified elements of the AAI standard would predict the actual relationship between varying ambient concentrations of oxides of nitrogen and sulfur and steady state ANC levels across the distribution of water bodies within the various ecoregions in the U.S. Because of this, there is considerable uncertainty as to the actual degree of protectiveness that such a standard would provide, especially for acid-sensitive ecoregions. The Administrator recognizes that the AAI equation, with factors quantified in the ranges discussed above and described more fully in the PA, generally performs well in identifying areas of the country that

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are sensitive to such acidifying deposition and indicates, as expected, that lower ambient levels of oxides of nitrogen and sulfur would lead to higher calculated AAI values. However, the uncertainties discussed here are critical for determining the actual degree of protection that would be afforded such areas by any specific target ANC level and percentile of water bodies that would be chosen in setting a new AAI-based standard, and thus for determining an appropriate AAI-based standard that meets the requirements of section 109.

In considering these uncertainties, the Administrator notes that CASAC acknowledged that important uncertainties remain that would benefit from further study and data collection efforts, which might lead to potential revisions or modifications to the form of the standard developed in the PA. She also notes that CASAC encouraged the Agency to engage in future monitoring and model evaluation efforts to help inform the specification of model-derived elements in the AAI equation.

Based on the above considerations, the Administrator has determined that it is not appropriate under section 109 to set a new multi-pollutant standard to address deposition-related effects of oxides of nitrogen and sulfur on aquatic acidification at this time. Setting a NAAQS generally involves consideration of the degree of uncertainties in the science and other information, such as gaps in the relevant data and, in this case, limitations in the evaluation of the application of relevant ecological and atmospheric models at an ecoregion scale. As noted above, the issue here is not a question of uncertainties about the scientific soundness of the structure of the AAI, but instead uncertainties in the quantification and representativeness of the elements of the AAI as they vary in ecoregions across the country. At present, these uncertainties prevent an understanding of the degree of protectiveness that would be afforded to various ecoregions across the country by a new standard defined in terms of a specific

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nationwide target ANC level and a specific percentile of water bodies for acid-sensitive ecoregions and thus prevent identification of an appropriate standard.. The Administrator has considered whether these uncertainties could be appropriately accounted for by choosing either a more or less protective target ANC level and percentile of water bodies than would otherwise be chosen if the uncertainties did not substantially limit the confidence that can appropriately be ascribed to the quantification of the AAI elements. However, in the Administrator's judgment, the uncertainties are of such nature and magnitude that there is no reasoned way to choose such a specific nationwide target ANC level or percentile of water bodies that would appropriately account for the uncertainties, since neither the direction nor the magnitude of change from the target level and percentile that would otherwise be chosen can reasonably be ascertained at this time.

Based on the above considerations, the Administrator judges that the current limitations in relevant data and the uncertainties associated with specifying the elements of the AAI based on modeled factors are of such nature and degree as to prevent her from reaching a reasoned decision such that she is adequately confident as to what level and form (in terms of a selected percentile) of such a standard would provide any particular intended degree of protection of public welfare that the Administrator determined satisfied the requirements to set an appropriate standard under section 109. While acknowledging that CASAC supported moving forward to establish the standard developed in the PA, the Administrator also observes that CASAC supported conducting further field studies that would better inform the continued development or modification of such a standard. Given the large uncertainties and complexities inherent in quantifying the elements of such a standard, largely deriving from the unprecedented nature of the standard under consideration in this review, and having fully considered CASAC's advice,

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the Administrator provisionally concludes that it is premature to set a new, multi-pollutant secondary standard for oxides of nitrogen and sulfur at this time, and as such she is proposing not to set such a new secondary standard.

While it is premature to set such a multi-pollutant standard at this time, the Administrator determines that the Agency should undertake a field pilot program to gather additional data, and that it is appropriate that such a program be undertaken before, rather than after, reaching a decision to set such a standard. As described below in section IV, the purpose of the program is to collect and analyze data so as to enhance our understanding of the degree of protectiveness that would likely be afforded by a standard based on the AAI as developed in the PA. This will provide additional information to aid the Agency in considering an appropriate multi-pollutant standard, specifically with respect to the acidifying effects of deposition of oxides of nitrogen and sulfur.PA. Data generated by this field program will also support development of an appropriate monitoring network that would work in concert with such a standard to result in the intended degree of protection. The data and analyses generated as a result of this program will serve to inform the next review of the NAAQS for oxides of nitrogen and sulfur. The information generated during the field program can also be used to help state agencies and EPA better understand how an AAI-based standard would work in terms of the implementation of such a standard.

Based on the above considerations, the Administrator is proposing not to set a new multi-pollutant AAI-based secondary standard for oxides of nitrogen and sulfur in this review. In reaching this decision, the Administrator recognizes that the new NO_2 and SO_2 primary 1-hour standards set in 2010, while not ecologically relevant for a secondary standard, will nonetheless result in reductions in oxides of nitrogen and sulfur that will directionally benefit the

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environment by reducing NOy and SOx deposition to sensitive ecosystems. EPA is proposing to revise the secondary standards by adding secondary standards identical to the NO_2 and SO_2 primary 1-hour standards set in 2010. More specifically, EPA is proposing a 1-hour secondary NO_2 standard set at a level of 100 ppb and a 1-hour secondary SO_2 standard set at a level of 75 ppb. While this will not add secondary standards of an ecologically relevant form to address deposition-related effects, it will directionally provide some degree of additional protection. This is consistent with the view that the current secondary standards are neither sufficiently protective nor appropriate in form, but that it is not appropriate to propose to set a new, ecologically relevant multi-pollutant secondary standard at this time, for all of the reasons discussed above.

While not a basis for this decision, the Administrator also recognizes that a new, innovative AAI-based standard would raise significant implementation issues that would need to be addressed consistent with the CAA requirements for implementation-related actions following the setting of a new NAAQS. It will take time to address these issues, during which the Agency will be conducting a field pilot program to gather relevant data and the environment will benefit from reductions in oxides of nitrogen and sulfur resulting from the new NO_2 and SO_2 primary standards, as noted above, as well as reductions expected to be achieved from EPA's Cross-State Air Pollution Rule and Mercury and Air Toxics standards. These implementation-related issues are discussed in more detail below in section IV.A.5.

The Administrator solicits comment on all aspects of this proposed decision, including the framework and elements of a multi-pollutant standard for oxides of nitrogen and sulfur to address deposition-related effects on sensitive ecosystems, with a focus on aquatic acidification, and the uncertainties and complexities associated with the development of such a standard at this

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time. The Administrator also solicits comment on the field pilot program and related monitoring methods as discussed below in section IV.

IV. Field Pilot Program and Ambient Monitoring

This section describes EPA's plans for a field pilot program and the evaluation of monitoring methods for ambient air indicators of NO_y and SO_x to implement the Administrator's decision to undertake such a field monitoring program in conjunction with her decision to propose not to set a new multi-pollutant secondary standard in this review, as discussed above in section III.H. As noted above and discussed below in section IV.A, the field pilot program is intended to collect and analyze data so as to enhance our understanding of the degree of protectiveness that would likely be afforded by a standard based on the AAI as developed in the PA. Data generated by this field program would also support development of an appropriate monitoring network that would work in concert with such a standard to result in the intended degree of protection. As discussed below in section IV.B, the evaluation of monitoring methods focuses on the development of Federal Reference Methods/Federal Equivalent Methods (FRM/FEM) for NO_y and SO_x. The EPA notes that the monitoring program described here is intended to be coordinated with EPA's CASTNET as a supplement to existing monitoring programs and is beyond the scope of the current CASTNET program.

A. Field Pilot Program

This section presents the objectives of a field pilot program (section IV.A.1) that would gather relevant field data over a 5-year period in a sample of three to five sensitive ecoregions across the country. An overview of the scope and structure of the field program, with a focus on measurements of ambient air indicators of oxides of nitrogen and sulfur, is presented in section IV.A.2. Section IV.A.3 explains the role of additional complementary measurements beyond the

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ambient air indicators that would be included in the program, and section IV.A.4 discusses a parallel longer-term research agenda, both of which are guided by the uncertainties discussed above in section III. Section IV.A.5 identifies implementation challenges presented by an AAI-based standard that could be addressed in parallel with a field pilot program. Section IV.A.6 discusses engagement with stakeholder groups as part of the planned pilot program.

1. Objectives

Consideration of a new multi-pollutant standard to address deposition-related effects on sensitive aquatic ecoregions raises unique challenges relative to those typically raised in reviews of existing NAAQS for which an established network of FRM/FEM monitors, designed to measure the indicator pollutant, is generally available. The primary goal of this field pilot program, and the related monitoring program discussed in section IV.B, is to enhance our understanding of the degree of protectiveness that would likely be afforded by a standard based on the AAI, as described above in section III, so as to aid the Agency in considering an appropriate multi-pollutant standard that would be requisite to protect public welfare consistent with section 109 of the CAA, through the following objectives:

(1) Evaluate measurement methods for the ambient air indicators of NO_y and SO_x and consider designation of such methods as FRMs;

(2) Examine the variability and improve characterization of concentration and deposition patterns of NO_y and SO_x, as well as reduced forms of nitrogen, within and across a number of sensitive ecoregions across the country;

(3) Develop updated ecoregion-specific factors (i.e., F1 through F4) for the AAI equation based in part on new observed air quality data within the sample ecoregions as well as on updated nationwide air quality model results and expanded critical load

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data bases, and explore alternative approaches for developing such representative factors;

(4) Calculate ecoregion-specific AAI values using observed NO_y and SO_x data and updated ecoregion-specific factors to examine the extent to which the sample ecoregions would meet a set of alternative AAI-based standards;

(5) Develop air monitoring network design criteria for an AAI-based standard;

(6) assess the use of total nitrate measurements as a potential alternative indicator for NO_y;

(7) Support related longer-term research efforts, including enhancements to and evaluation of modeled dry deposition algorithms; and

(8) Facilitate stakeholder engagement in addressing implementation issues associated with possible future adoption of an AAI-based standard.

2. Overview of Field Pilot Program

The CASTNET program (Figure IV-1) affords an available infrastructure relevant to an AAI-based standard, given the location of sites in some acid-sensitive ecoregions and various measurements of sulfur and nitrogen species. The EPA plans to use CASTNET sites in selected acid-sensitive ecoregions to serve as the platform for this pilot program, potentially starting in late 2012 and extending through 2018. The CASTNET sites in three to five ecoregions in acid-sensitive areas would collect NO_y and SO_x (i.e., SO_2 and p-SO_4) measurements over a 5-year period. The initial step in developing a data base of observed ambient air indicators for oxides of nitrogen and sulfur requires the addition of NO_y samplers at the pilot study sites so that a full complement of indicator measurements are available to calculate AAI values. These CASTNET

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sites would also be used to make supplemental observations useful for evaluation of CMAQ's characterization of factors F2 –F4 in the AAI equation.

The selected ecoregions would account for geographic variability by including regions from across the U.S., including the east, upper midwest and west. Each selected region would have at least two existing CASTNET sites. Each of the pilot CASTNET sites would be used to evaluate the performance of the established methods, data retrieval and reporting procedures used in the AAI equation.

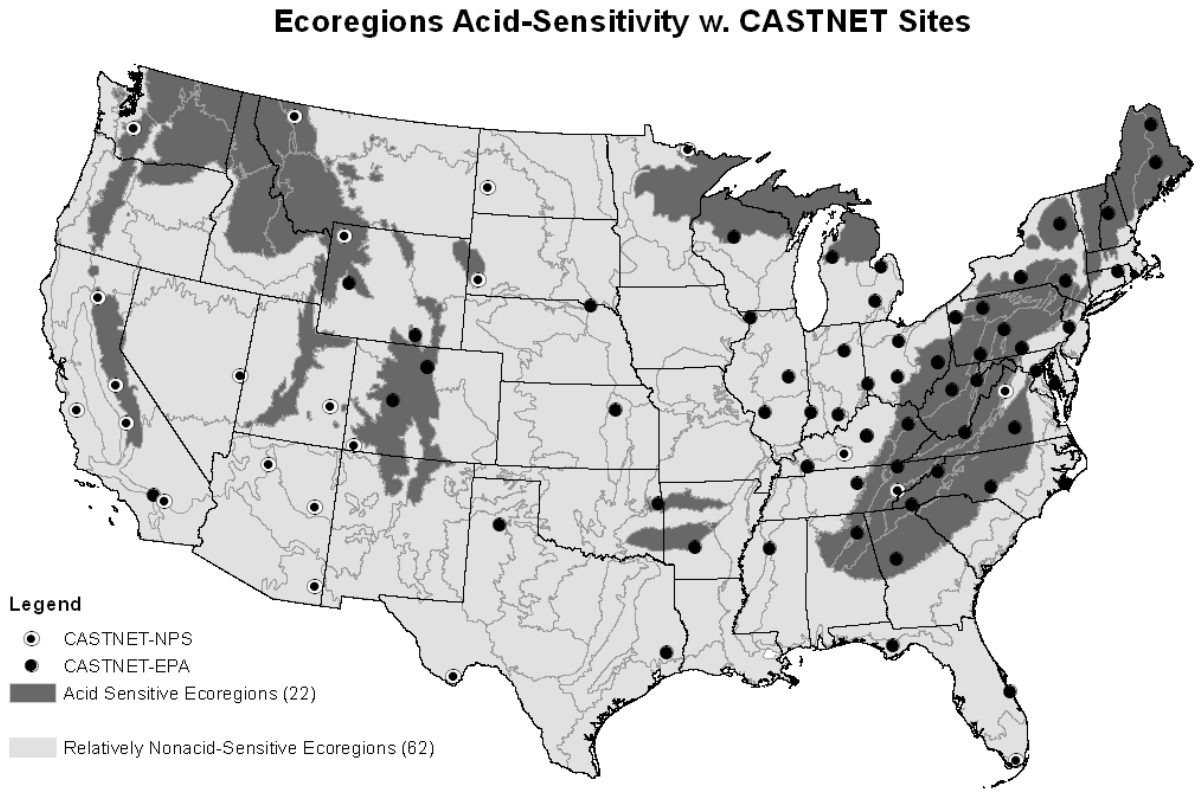

Figure IV-1. Location of CASTNET sites in relation to acid sensitive ecoregions.

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Over the course of this 5-year pilot program, the most current national air quality modeling, based on the most current national emissions inventory, would be used to develop an updated set of F2 – F4 factors. A parallel multi-agency national critical load data base development effort would be used as the basis for calculating updated F1 factors. As discussed above in section III.B, these factors would be based on average parameter values across an ecoregion. Using this new set of F factors, observations of NO_y and SO_x derived from the pilot program, averaged across each ecoregion, would be used to calculate AAI values in the sample ecoregions. The data from the pilot program would also be used to examine alternative approaches to generating representative air quality values, such as examining the appropriateness of spatial averaging in areas of high spatial variability.

3. Complementary Measurements

Complementary measurements may be performed at some sites in the pilot network to reduce uncertainties in the recommended methods and better characterize model performance and application to the AAI. The CASAC Air Monitoring and Methods Subcommittee (AMMS) advised EPA that such supplemental measurements were of critical importance in a field measurement program related to an AAI-based standard (Russell and Samet, 2011b).

Candidate complementary measurements to address sulfur, in addition to those provided by the CASTNET filter pack (CFP), include trace gas continuous SO_2 and speciated $PM_{2.5}$ measurements. The co-located deployment of a continuous SO_2 analyzer with the CFP for SO_2 will provide test data for determining suitability of continuous SO_2 measurements as a Federal Equivalent Method (FEM), as well as producing valuable time series data for model evaluation purposes. The weekly averaging time provided by the CFP adequately addresses the annual-

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average basis of an AAI-based secondary standard, but would not be applicable to short-term (i.e., 1-hour) averages associated with the primary SO_2 standard. Conversely, because of the low concentrations associated with many acid-sensitive ecoregions, existing SO_2 Federal Reference Methods (FRMs) designated for use in determining compliance with the primary standard would not necessarily be appropriate for use in conjunction with an AAI-based secondary standard.

Co-locating the $PM_{2.5}$ sampler used in the EPA Chemical Speciation Network and the Interagency Monitoring of Protected Visual Environments (IMPROVE) network at pilot network sites would allow for characterizing the relationship between the CFP-derived p-SO_4 and the speciation samplers used throughout the state and local air quality networks. Note that CASTNET already has several co-located IMPROVE chemical speciation samplers. Because the AAI equation is based on concentration of p-SO_4, the original motivation for capturing all particle size fractions is not as important relative to simply capturing the concentration of total p-SO_4.

Candidate measurements to complement oxidized nitrogen measurements, in addition to the CFP, include a mix of continuous and periodic sampling for the dominant NO_y species, namely NO, true NO_2, PAN, HNO_3, and p-NO_3. While there are several approaches to acquiring these measurements, perhaps the most efficient strategy would take advantage of the available CFP for total nitrate, and add a three-channel chemiluminescence instrument that will cycle between NO_y, true NO_2 and NO by adding photolytic detection for true NO_2. Other options for measuring true NO_2 would include adding either a stand-alone photolytic or cavity ring-down spectroscopy instrument. Measurements of PAN may be acquired either on a periodic basis through canister sampling and subsequent laboratory analysis or through emerging in-situ sampling and analysis methods. Although the CFP yields a reliable measurement of total nitrate,

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the t-NO_3 (i.e., the sum of HNO_3 and p-NH_4) value, strong consideration may be given to direct measurement of HNO_3, which has the highest deposition velocity of all the dominant NO_y species. Similar to the use of continuous SO_2 data, these speciated NO_y data serve two purposes: evaluating total NO_y instrument behavior and evaluating air quality models. The measurement of individual NO_y species can be used to generate site-specific NOy values for comparison to modeled NO_y, and will likely provide insight into and improvement of modeled dry deposition.

The CASAC AMMS (Russell and Samet, 2011b) recommended that EPA consider the use of t-NO_3 obtained from CASTNET sampling as an indicator for NO_y, reasoning that t-NO_3 is typically a significant fraction of deposited oxidized nitrogen in rural environments and CASTNET measurements are widely available. Collection of this data would support further consideration of using the CFP for t-NO_3 as the indicator of oxides of nitrogen for use in an AAI-based secondary standard.

The CASAC AMMS also recommended that total NH_x (NH_3 and p-NH_4) be considered as a proxy for reduced nitrogen species, reasoning that the subsequent partitioning to NH_3 and p-NH_4 may be estimated using equilibrium chemistry calculations. Reduced nitrogen measurements are used to evaluate air quality modeling which is used in generating factor F2. Additional studies are needed to determine the applicability of NH_x measurements and calculated values of NH_3 and NH_4 to the AAI.

The additional supplemental measurements of speciated NO_y, continuous SO_2 and NH_x will be used in future air quality modeling evaluation efforts. Because there often is significant lag in the availability of contemporary emissions data to drive air quality modeling, the complete use of these data sets will extend beyond the 5-year collection period of the pilot program. Consequently, the immediate application of those data will address instrument performance

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comparisons that explore the feasibility of using continuous SO_2 instruments in rural environments, and using the speciated NO_y data to assess NO_y instrument performance. Although contemporary air quality modeling will lag behind measurement data availability, the observations can be used in deposition models to compare observed transference ratios with the previously calculated transference ratios to test temporal stability of the ratios.

An extended water quality sampling effort should parallel the air quality measurement program to address some of the uncertainties related to factor F1 and the representativeness of the nth percentile critical load as discussed in section III.B.5.b.i. The objective of the water quality sampling would be to develop a larger data base of critical loads in each of the pilot ecoregions such that the nth percentile can adequately be characterized in terms of representing all water bodies. Opportunities to leverage and perhaps enhance existing ecosystem modeling efforts enabling more advanced critical load modeling and improved methods to estimate base cation production would be pursued. For example, areas with ongoing research studies producing data for dynamic critical load modeling would be considered when selecting the pilot ecoregions.

4. Complementary areas of research

The EPA recognizes that a source of uncertainty in an AAI-based secondary standard that would not be directly addressed in the pilot program stems from the uncertainty in the model used to link atmospheric concentrations to dry deposition fluxes. Currently, there are no ongoing direct dry deposition measurement studies at CASTNET sites that can be used to evaluate modeled results. It was strongly recommended by CASAC AMMS that a comprehensive sampling-intensive study be conducted in at least one, preferably two sites in different ecoregions to assess characterization of dry deposition of sulfur and nitrogen. These sites would

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be the same as those for the complementary measurements described above, but they would afford an opportunity to also complement dry deposition process research that benefits from the ambient air measurements collected in the pilot program. The concerns regarding uncertainties underlying an AAI-based secondary standard suggest that research that includes dry deposition measurements and evaluation of dry deposition models should be a high priority.

Similar leveraging should be pursued with respect to ecosystem research activities. For example, studies that capture a suite of soil, vegetation, hydrological, and water quality properties that can help evaluate more advanced critical load models would complement the atmospheric-based pilot program. In concept, such studies could provide the infrastructure for true multi-pollutant, multi-media "super" sites assuming the planning, coordination, and resource facets can be aligned. While this discussion emphasizes the opportunity of leveraging ongoing research efforts, consideration could be given to explicitly including related research components directly in the pilot program.

5. Implementation challenges

The CAA requires that once a NAAQS is established, designation and implementation must move forward. With a standard as innovative as the AAI-based standard considered in this review, the Administrator believes that its success will be greatly improved if, while additional data are being collected to reduce the uncertainties discussed above, the implementing agencies and other stakeholders have an opportunity to discuss and thoroughly understand how such a standard would work. And since, as noted above, emissions reductions that are directionally correct to reduce aquatic acidification will be occurring as a result of other CAA programs, the Administrator believes that this period of further discussion will not delay progress but will ensure that once implementation is triggered, agencies will be prepared to implement it

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successfully.

Consideration of an AAI-based secondary standard for oxides of nitrogen and sulfur would present significant implementation challenges because it involves multiple, regionally-dispersed pollutants and relatively complex compliance determinations based on regionally variable levels of NOy and SOx concentrations that would be necessary to achieve a national ANC target.. The anticipated implementation challenges fall into three main categories: monitoring and compliance determinations for area designations, pre-construction permit application analyses of individual source impacts, and State Implementation Plan (SIP) development. Several overarching implementation questions that we anticipate will be addressed in parallel with the field pilot program's five-year data collection period include:

(1) What are the appropriate monitoring network density and siting requirements to support a compliance system based on ecoregions?

(2) Given the unique spatial nature of the secondary standard (e.g., ecoregions), what are the appropriate parameters for establishing nonattainment areas?

(3) How can new or modified major sources of oxides of nitrogen and oxides of sulfur emissions assess their ambient impacts on the standard and demonstrate that they are not causing or contributing to a violation of the NAAQS for preconstruction permitting? To what extent does the fact that a single source may be impacting multiple areas, with different acid sensitivities and variable levels of NOy and SOx concentrations that would be necessary to achieve a national ANC target, complicate this assessment and how can these additional complexities best be addressed?

(4) What additional tools, information, and planning structures are needed to assist states with SIP development, including the assessment of interstate pollutant transport and

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deposition?

(5) Would transportation conformity apply in nonattainment and maintenance areas for this secondary standard, and, if it does, would satisfying requirements that apply for related primary standards (e.g., ozone, $PM_{2.5}$, and NO_2) be demonstrated to satisfy requirements for this secondary standard?

6. Final monitoring plan development and stakeholder participation

The existing CASTNET sampling site infrastructure provides an effective means of quickly and efficiently deploying a monitoring program to support potential implementation of an AAI-based secondary standard, and also provides an additional opportunity for federally managed networks to collaborate and support the states, local agencies and tribes (SLT) in determining compliance with a secondary standard. A collaborative effort would help to optimize limited federal and SLT monitoring funds and would be beneficial to all involved. The CASTNET is already a stakeholder-based program with over 20 participants and contributors, including federal, state and tribal partners.

The CASAC AMMS generally endorsed the technical approaches used in CASTNET, but concerns were raised by individual representatives of state agencies concerning the perception of EPA-controlled management aspects of CASTNET and data ownership. Potential approaches to resolve these issues will be developed and evaluated in existing National Association of Clean Air Agencies (NACAA)/EPA ambient air monitoring workgroups. The EPA Office of Air and Radiation (which includes the Office of Air Quality Planning Standards, OAQPS; and the Office of Atmospheric Program's Clean Air Markets Division, OAP-CAMD), and their partners on the NACAA monitor steering committee will develop a prioritized specific plan that identifies the three to five ecoregions and the instrumentation to be deployed. The EPA anticipates that a cost

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estimate of the plan with priorities and options will be developed by January, 2012. Although this pilot program is focused on data collection, the plan will include details of the data analysis approaches as well as a vehicle that incorporates engagement from those within EPA and SLTs to foster progress on the implementation questions noted above in section IV.A.5.

If an AAI-based secondary standard were to be set in the future, deployment of a full national network would follow the pilot monitoring program. The number of sites deployed in the network will lead to increased confidence in capturing spatial patterns of air quality. Recognizing that this section presents the general elements of the field pilot programs, EPA intends to develop a more detailed field pilot program plan through a process that will engage the air quality management and research (atmospheric and ecosystem) communities, as well as other federal agencies, state and local agencies, and non-government based centers of expertise. The EPA is seeking comment and input on all aspects of this field pilot program.

.B. Evaluation of Monitoring Methods

The EPA generally relies on monitoring methods that have been designated as FRMs or FEMs for the purpose of determining the attainment status of areas with regard to existing NAAQS. Such FRMs or FEMs are generally required to measure the air quality indicators that are compared to the level of a standard to assess compliance with a NAAQS. Prior to their designation by EPA as FRM/FEMs through a rulemaking process, these methods must be determined to be applicable for routine field use and need to have been experimentally validated by meeting or exceeding specific accuracy, reproducibility, and reliability criteria established by EPA for this purpose. As discussed above in section III.A, the ambient air indicators being considered for use in an AAI-based standard include SO_2, particulate sulfate (p-SO_4), and total reactive oxides of nitrogen (NO_y).

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The CASTNET provides a well established infrastructure that would meet the basic location and measurement requirements of an AAI-based secondary standard given the rural placement of sites in acid sensitive areas. In addition, CFPs currently provide very economical weekly, integrated average concentration measurements of SO_2, p-SO_4, ammonium ion (NH_4) and t-NO_3, the sum of HNO_3 and p-NO_3.

While routinely operated instruments that measure SO_2, p-SO_4, NO_y and/or t-NO_3 exist, instruments that measure p-SO_4, NO_y, t-NO_3, or the CFP for SO_2 have not been designated by EPA as FRMs or FEMs. The EPA's Office of Research and Development has initiated work that will support future FRM designations by EPA for SO_2 and p-SO_4 measurements based on the CFP. Such a designation by EPA could be done for the purpose of facilitating consistent research related to an AAI-based standard and/or in conjunction with setting and supporting an AAI-based secondary standard.

Based on extensive review of literature and available data, the EPA has identified potential methods that appear suitable for measuring each of the three components of the indicators. These three methods are being considered as new FRMs to be used for measuring the ambient concentrations of the three components that would be needed to determine compliance with an AAI-based secondary standard.

For the SO_2 and p-SO_4 measurements, EPA is considering the CFP method, which provides weekly average concentration measurements for SO_2 and p-SO_4. This method has been used in the EPA's CASTNET monitoring network for 15 years, and strongly indicates that it will meet the requirements for use as an FRM for the SO_2 and p-SO_4 concentrations for an AAI-based secondary standard.

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Although the CFP method would provide measurements of both the SO_2 and p-SO_4 components in a unified sampling and analysis procedure, individual FRMs will be considered for each. The EPA recognizes that an existing FRM to measure SO_2 concentrations using ultra-violet fluorescence (UVF) exists (40 CFR Part 50, Appendix A-1) for the purpose of monitoring compliance for the primary SO_2 NAAQS. However, several factors suggest that the CFP method would be superior to that UVF FRM for monitoring compliance with an AAI-based secondary standard and will be discussed in more detail below.

For monitoring the NO_y component, a continuous analyzer for measuring NO_y is commercially available and is considered to be suitable for use as an FRM. This method is similar in design to the existing NO_2 FRM (described in 40 CFR Part 50, Appendix F), which is based on the ozone chemiluminescence measurement technique. The method is adapted to and further optimized to measure all NO_y. However, this NO_y method requires further evaluation before it can be fully confirmed as a suitable FRM. The EPA is currently completing a full scientific assessment of the NO_y method to determine whether it would be appropriate to consider for designation by EPA as an FRM. Specific details on these three methods are given below.

On February 16, 2011, EPA presented this set of potential FRMs to the CASAC AMMS for their consideration and comment. In response, the CASAC AMMS stated that, overall, it believes that EPA's planned evaluation of methods for measuring NO_y, SO_2 and p-SO_4 as ambient air indicators is a suitable approach in concept. On supporting the CFP method as a potential FRM for SO_2, CASAC stated that they felt that the CFP is adequate for measuring long-term average SO_2 gas concentrations in rural areas with low levels (less than 5 parts per billion by volume (ppbv)) and is therefore suitable for consideration as an FRM. For p-SO_4,

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CASAC generally supports the use of the CFP as a potential FRM for measuring p-SO_4 for an AAI-based secondary standard. The method has been relatively well-characterized and evaluated, and it has a documented, long-term track record of successful use in a field network designed to assess spatial patterns and long-term trends.

On supporting the photometric NO_y method as a potential FRM, CASAC concluded that the existing NOy method is generally an appropriate approach for the indicator. However, CASAC agrees that additional characterization and research is needed to fully understand the method in order to designate it as a FRM. The EPA is now soliciting public comment on these methods as to their adequacy, suitability, and relative merits as FRMs for purposes of monitoring to determine compliance with an AAI-based secondary standard.

1. Potential FRMs for SO_2 and p-SO_4

The CFP is a combined, integrated sampling and analysis method based on the well-established measurement technology that has been used extensively in EPA's CASTNET monitoring network (see http://www.epa.gov/castnet). This method is in current use at over 80 monitoring sites and has been in use at not less than 40 sites for over 15 years. This method employs a relatively simple and inexpensive sampler and uses four 47-mm filters placed in an open-faced filter pack to simultaneously collect integrated filter samples for the SO_2 and p-SO_4 components. In addition, the CFP is also capable of the collection of t-NO_3, the sum of HNO_3 and p-NO_3.

The first stage of the filter pack assembly contains a Teflon® filter that collects p-SO_4^{2-} and p-NO_3, the second stage contains a nylon filter that collects SO_2 (as SO_4^{2-}) and HNO_3, and the third stage contains two cellulose fiber filters impregnated with potassium carbonate (K_2CO_3) that collect any remaining SO_2 (as SO_4^{2-}). The sampler collects 1-week integrated samples at a

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very low, controlled flow rate (1.5 or 3 L/min) in an attempt simulate actual deposition. Weekly averaged SO_2 and p-SO_4 concentrations could then be averaged over a 1-year period to calculate annual average values.

Upon sample completion, the species-specific filters are extracted, with subsequent analysis by the well-established and documented ion chromatographic (IC) analytical technique. During the IC analysis, an aliquot of a filter extract is injected into a stream of eluent (ion chromatography mobile phase, generally a millimolar-strength solution of carbonate-bicarbonate) and passed through a series of ion exchangers. The anions of interest are separated on the basis of their relative affinities for a low capacity and the strongly basic anion exchanger (guard and separator column). The separated anions are directed onto a cation exchanger (suppressor column) where they are converted to their highly conductive acid form, and the eluent is converted to a weakly conductive form. The now-separated anions, each in their acid form, are measured by conductivity. They are identified on the basis of retention time compared to that of standards and quantified by measurement of peak area compared to the peak areas of calibration standards.

Calibration and quality assurance for the method are applied to the sample filters, the analytical processes, and the flow rate measurement and control aspects of the sampler. Overall method performance is typically assessed with collocated samplers. These quality assurance techniques are routinely used and have proved adequate for other types of FRMs and equivalent methods in air monitoring network service.

The measurement and analytical procedures and past performance data associated with the CFP method are well documented and available through Quality Assurance Performance Plans (QAPPs), Standard Operating Procedures (SOPs) and annual reports (US EPA, 2010a and

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2010b). The accumulated database on the CFP method is substantial and indicates that the method is sound, stable and has good reliability in routine, field operation. Data quality assessment results show the method to have good reproducibility, with collocated and analytical precision values in the range of 2 percent to 10 percent (excluding very low concentration measurements near the method detection limits; US EPA 2010b).

Data quality objectives (DQOs) for a new FRM would be based upon current DQOs being used for this method by EPA's OAP/CAMD and the NPS, the federal managers of CASTNET (US EPA, 2010a). In its current state, the CFP method is expected to meet or exceed (as past CASTNET data have indicated; US EPA, 2010b) the expected FRM DQOs, even when deployed in new monitoring networks outside of CASTNET. In addition, CASTNET samples have agreed favorably with other measures of SO_2 and p-SO_4 in comparison studies. For example, in direct comparison with an annular denuder sampler (ADS) method, CASTNET/ADS ratios for SO_2 and p-SO_4 were generally on the order of 0.9-1.1 (Lavery et al, 2009; Sickles et al, 1999; Sickles et al, 2008), thus illustrating the accuracy of the CFP method in the determination of long-term average SO_2 and p-SO_4 concentrations. The EPA believes that the CFP method would be fully adequate as an FRM in determining yearly average SO_2 and p-SO_4 concentrations for compliance determination purposes.

The EPA recognizes that an existing FRM for SO_2 has proven adequate for the purposes of monitoring compliance for the primary SO_2 NAAQS, specifically the newly-promulgated 1-hour standard. However, this FRM is better suited to the shorter-term, higher concentration primary and secondary SO_2 NAAQS, and there is substantial uncertainty as to the adequacy of this SO_2 FRM for monitoring the lower concentrations relevant to determining compliance with an AAI-based secondary standard. The performance specifications for SO_2 FRM analyzers (40

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CFR Part 53, Table B-1) require a lower detectable limit (LDL) of 0.002 ppm for the standard measurement range and 0.001 ppm for the lower measurement range. These requirements correspond to mass per unit volume concentrations of 5.24 and 2.62 $\mu g/m^3$, respectively. Analysis of 2009 CASTNET data shows that of the 84 CASTNET sampling sites, 63 measured annual average SO_2 concentrations below even the lower of these LDL requirements of 2.62 $\mu g/m^3$ for the lower range SO_2 FRM (US EPA, 2010a). In addition, 11 of the 84 sites measured annual (2009) average SO_2 concentrations very near or below the manufacturers' reported detection limits for trace level UVF SO_2 monitors. Further, it is likely that the number of sites with annual average SO_2 concentration below both the SO_2 FRM LDL and the manufacturers reported detection limits will increase due to expected declines in mean SO_2 concentrations (US EPA, 2010b). For these reasons, EPA is considering the CFP method for use as the FRM for monitoring the SO_2 component of an ambient air indicator for oxides of sulfur, with a recommendation for additional study and data collection to evaluate further the possible applicability of the continuous UVF SO_2 FRM for this purpose.

2. Potential FRM for NO_y

Atmospheric concentrations of NO_y are measured continuously by an analyzer that photometrically measures the light intensity, at wavelengths greater than 600 nanometers (nm), resulting from the chemiluminescent reaction of ozone (O_3) with NO in sampled air. This method is very similar to the chemiluminescence NO/NO_2 analyzers widely used to collect NO_2 monitoring data for determining compliance with the NO_2 NAAQS. The various oxides of nitrogen species, excluding NO, are first quantitatively reduced to NO by means of a catalytic converter. These species include NO_2, HNO_2, PANs, HNO_3 and p-NO_3. The NO, which commonly exists in ambient air, passes through the converter unchanged, and, when combined

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with the NO resulting from the catalytic conversion of the other oxides of nitrogen, a measurement of the total NO_y concentration results. To maximize the conversion of the more chemically active oxides of nitrogen species, the converter is located externally, at or near the air sample inlet probe. This location minimizes losses of these active species that could otherwise occur from chemical reactions and wall losses in the sample inlet line.

The NO_y analyzer is a suitable, commercially produced continuous chemiluminescence analyzer that includes an ozone generator, a reaction cell, a photometric detector, wavelength filters as necessary to reduce sensitivity to wavelengths below 600 nanometer (nm), a pump and flow control system to draw atmospheric air through the converter and into the reaction cell, a suitable converter, a system to control the operation of the analyzer, and appropriate electronics to process and quantitatively scale the photometric signals. The converter contains a catalyst such as molybdenum and is heated to an optimum temperature designed to optimize the conversion of the various oxides of nitrogen to NO. It is connected to the analyzer via suitable lengths of Teflon® tubing. Hourly NO_y measurements obtained by the analyzer would be averaged over the same 7-day period used by the CFP method to measure the SO_2 and p-SO_4 components, with further averaging over a 1-year period.

Commercial NO_y analyzers are currently available, and the analyzers have been used for a variety of monitoring applications. During the 2006 TexAQS Radical and Aerosol Measurement Project (TRAMP), Luke et al., 2010, compared measured NO_y concentrations obtained with an NO_y instrument based upon the above mentioned methodology with the sum of measured individual NO_y species (i.e., NOy*i* = $NO+NO_2+HNO_3+PANs+HNO_2+p\text{-}NO_3$). This comparison yielded excellent overall agreement during both day ([NOy](ppb) = [NOy*i*](ppb) × 1.03 – 0.42; $r^2 = 0.9933$) and night time ([NOy](ppb) = [NOy*i*](ppb) × 1.01 – 0.18; $r^2 = 0.9975$)

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periods (Luke *et al*, 2010). The results of this study show that this NO_y method is capable of the accurate determination of all the atmospherically relevant NO_y components, resulting in an accurate determination of total NO_y concentrations. The NO_y instruments have been routinely operated in networks such as SouthEastern Aerosol Research and Characterization (SEARCH), dating back several years. In addition, state monitoring agencies across the U.S. have begun, starting in 2009, the routine operation of commercially available NO_y instrumentation in anticipation of EPA's NCore network transitioning to full operation in 2011.

These initial assessments described above are promising and indicate that the photometric NO_y method appears to be accurate, reliable, and capable of routine network operation. As a result, the method is most likely capable for use as an FRM for determining atmospheric NO_y concentrations as a component in determining compliance with an AAI-based secondary standard. However, as described below, this continuous method for NO_y requires additional time for further evaluation before it can be fully confirmed for adoption as a FRM. The EPA has identified measurement uncertainties and some remaining science questions associated with this method. Among these are: (a) the ability of the method to capture all components of NO_y relevant to nitrogen deposition, (b) the efficiency of the molybdenum converter in converting all oxides of nitrogen to NO for detection (excluding NO_2, as this conversion is already well documented), (c) appropriate inlet height specifications to minimize any bias associated with vertical concentration gradients of key NO_y components, (d) identification and quantification of potential measurement interferences in the NO_y determination, and (e) development and demonstration of effective calibration/challenge procedures to best represent the various mixtures of NO_y components that are expected to be present in the different air sheds across the U.S.

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To address these NO_y method uncertainties and to fully assess this method for use as the NO_y FRM, EPA has developed a detailed research plan (Russell and Samet, 2011b) which was presented to the CASAC AMMS on February 16, 2011. In response, CASAC recognized the need for, and supported the general outline of EPA's research plan to evaluate the NO_y method for potential designation as an FRM (US EPA, 2011). In addition, the CASAC AMMS suggested additional areas of research associated with the photometric NO_y method that warrant further assessment prior to final designation of the method as the NO_y FRM. These include operation of the method during extremely low temperature conditions to investigate possible condensation in sample lines, method detection limits relative to low levels expected in remote areas, and ambient-based method evaluations in various air sheds across the U.S. In response to these CASAC AMMS suggestions, EPA is carrying out studies, in addition to the tasks outlined in the research plan, for the NO_y method. The results of these studies will likely take a year or more to become available. As noted previously, EPA anticipates that these results will be favorable and will confirm the adequacy of the NO_y method as a suitable FRM for determining compliance with an AAI-based secondary standard.

V. Statutory and Executive Order Reviews

A. Executive Order 12866: Regulatory Planning and Review and Review and Executive Order 13563: Improving Regulation and Regulatory Review

Under Executive Order 12866 (58 FR 51735, October 4, 1993), this action is a "significant regulatory action." Accordingly, EPA submitted this action to the Office of Management and Budget (OMB) for review under Executive Orders 12866 and 13563 (76 FR 3821, January 21, 2011), and any changes made in response to OMB recommendations have been documented in the docket for this action.

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B. Paperwork Reduction Act

This action does not impose an information collection burden under the provisions of the Paperwork Reduction Act, 44 U.S.C. 3501 et seq. Burden is defined at 5 CFR 1320.3(b). There are no information collection requirements directly associated with the establishment of a NAAQS under section 109 of the CAA.

C. Regulatory Flexibility Act

For purposes of assessing the impacts of today's rule on small entities, small entity is defined as: (1) a small business that is a small industrial entity as defined by the Small Business Administration's (SBA) regulations at 13 CFR 121.201; (2) a small governmental jurisdiction that is a government of a city, county, town, school district or special district with a population of less than 50,000; and (3) a small organization that is any not-for-profit enterprise which is independently owned and operated and is not dominant in its field.

After considering the economic impacts of today's proposed rule on small entities, I certify that this action will not have a significant economic impact on a substantial number of small entities. This proposed rule will not impose any requirements on small entities. Rather, this rule establishes national standards for allowable concentrations of oxides of nitrogen and sulfur in ambient air as required by section 109 of the CAA. See also American Trucking Associations v. EPA. 175 F. 3d at 1044-45 (NAAQS do not have significant impacts upon small entities because NAAQS themselves impose no regulations upon small entities). We continue to be interested in the potential impacts of the proposed rule on small entities and welcome comments on issues related to such impacts.

D. Unfunded Mandates Reform Act

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Title II of the Unfunded Mandates Reform Act of 1995 (UMRA), Public Law 104-4, establishes requirements for Federal agencies to assess the effects of their regulatory actions on State, local, and Tribal governments and the private sector. Under section 202 of the UMRA, EPA generally must prepare a written statement, including a cost-benefit analysis, for proposed and final rules with "Federal mandates" that may result in expenditures to state, local, and tribal governments, in the aggregate, or to the private sector, of $100 million or more in any 1 year. Before promulgating an EPA rule for which a written statement is needed, section 205 of the UMRA generally requires EPA to identify and consider a reasonable number of regulatory alternatives and to adopt the least costly, most cost-effective or least burdensome alternative that achieves the objectives of the rule. The provisions of section 205 do not apply when they are inconsistent with applicable law. Moreover, section 205 allows EPA to adopt an alternative other than the least costly, most cost-effective or least burdensome alternative if the Administrator publishes with the final rule an explanation why that alternative was not adopted. Before EPA establishes any regulatory requirements that may significantly or uniquely affect small governments, including tribal governments, it must have developed under section 203 of the UMRA a small government agency plan. The plan must provide for notifying potentially affected small governments, enabling officials of affected small governments to have meaningful and timely input in the development of EPA regulatory proposals with significant Federal intergovernmental mandates, and informing, educating, and advising small governments on compliance with the regulatory requirements.

This action contains no Federal mandates under the provisions of Title II of the Unfunded Mandates Reform Act of 1995 (UMRA), 2 U.S.C. 1531-1538 for state, local, or tribal governments or the private sector. Therefore, this action is not subject to the requirements of

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sections 202 or 205. Furthermore, as indicated previously, in setting a NAAQS EPA cannot consider the economic or technological feasibility of attaining ambient air quality standards; although such factors may be considered to a degree in the development of state plans to implement the standards. See also American Trucking Associations v. EPA, 175 F. 3d at 1043 (noting that because EPA is precluded from considering costs of implementation in establishing NAAQS, preparation of a Regulatory Impact Analysis pursuant to the Unfunded Mandates Reform Act would not furnish any information which the court could consider in reviewing the NAAQS). Accordingly, EPA has determined that the provisions of sections 202, 203, and 205 of the UMRA do not apply to this proposed decision. The EPA acknowledges, however, that any corresponding revisions to associated state implementation plan (SIP) requirements and air quality surveillance requirements, 40 CFR part 51 and 40 CFR part 58, respectively, might result in such effects. Accordingly, EPA will address, as appropriate, unfunded mandates if and when it proposes any revisions to 40 CFR parts 51 or 58.

E. Executive Order 13132: Federalism

This proposed rule does not have federalism implications. It will not have substantial direct effects on the states, on the relationship between the national government and the states, or on the distribution of power and responsibilities among the various levels of government, as specified in Executive Order 13132 because it does not contain legally binding requirements. Thus, the requirements of Executive Order 13132 do not apply to this rule.

EPA believes, however, that this proposed rule may be of significant interest to state governments. As also noted in section E (above) on UMRA, EPA recognizes that states will have a substantial interest in this rule and any corresponding revisions to associated SIP requirements and air quality surveillance requirements, 40 CFR part 51 and 40 CFR part 58,

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respectively. Therefore, in the spirit of Executive Order 13132 and consistent with EPA policy to promote communications between EPA and state and local governments, EPA specifically solicits comment on this proposed rule from state and local officials.

F. Executive Order 13175: Consultation and Coordination with Indian Tribal Governments

Executive Order 13175, entitled "Consultation and Coordination with Indian Tribal Governments" (65 FR 67249, November 9, 2000), requires EPA to develop an accountable process to ensure "meaningful and timely input by tribal officials in the development of regulatory policies that have tribal implications." This rule concerns the establishment of national standards to address the public welfare effects of oxides of nitrogen and sulfur.

This action does not have Tribal implications, as specified in Executive Order 13175 (65 FR 67249, November 9, 2000). It does not have a substantial direct effect on one or more Indian tribes, since tribes are not obligated to adopt or implement any NAAQS. Thus, Executive Order 13175 does not apply to this rule.

G. Executive Order 13045: Protection of Children from Environmental Health & Safety Risks

This action is not subject to EO 13045 because it is not an economically significant rule as defined in EO 12866.

H. Executive Order 13211: Actions that Significantly Affect Energy Supply, Distribution or Use

This action is not a "significant energy action" as defined in Executive Order 13211 (66 FR 28355, May 22, 2001), because it is not likely to have a significant adverse effect on the supply, distribution, or use of energy. This action concerns the establishment of national standards to address the public welfare effects of oxides of nitrogen and sulfur. This action does

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not prescribe specific pollution control strategies by which these ambient standards will be met. Such strategies will be developed by states on a case-by-case basis, and EPA cannot predict whether the control options selected by states will include regulations on energy suppliers, distributors, or users.

I. National Technology Transfer and Advancement Act

Section 12(d) of the National Technology Transfer and Advancement Act of 1995 (NTTAA), Public Law No. 104-113, 12(d) (15 U.S.C. 272 note) directs EPA to use voluntary consensus standards in its regulatory activities unless to do so would be inconsistent with applicable law or otherwise impractical. Voluntary consensus standards are technical standards (e.g., materials specifications, test methods, sampling procedures, and business practices) that are developed or adopted by voluntary consensus standards bodies. The NTTAA directs EPA to provide Congress, through OMB, explanations when the Agency decides not to use available and applicable voluntary consensus standards.

The EPA is not aware of any voluntary consensus standards that are relevant to the provisions of this proposed rule. The EPA welcomes any feedback on such standards that may be applicable.

J. Executive Order 12898: Federal Actions to Address Environmental Justice in Minority Populations and Low-Income Populations

Executive Order 12898 (59 FR 7629 (Feb. 16, 1994)) establishes federal executive policy on environmental justice. Its main provision directs federal agencies, to the greatest extent practicable and permitted by law, to make environmental justice part of their mission by identifying and addressing, as appropriate, disproportionately high and adverse human health or

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environmental effects of their programs, policies, and activities on minority populations and low-income populations in the United States.

EPA has determined that this proposed rule will not have disproportionately high and adverse human health or environmental effects on minority or low-income populations because it retains the level of environmental protection for all affected populations without having any disproportionately high and adverse human health or environmental effects on any population, including any minority or low-income population.

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List of Subjects in 40 CFR Part 50

Environmental protection, Air pollution control, Carbon monoxide, Lead, Nitrogen dioxide, Ozone, Particulater matter, Sulfur oxides.

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Dated: July 12, 2011

Lisa P. Jackson,

Administrator

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For the reasons set forth in the preamble, part 50 of chapter 1 of title 40 of the code of Federal regulations is proposed to be amended as follows:

PART 50-NATIONAL PRIMARY AND SECONDARY AMBIENT AIR QUALITY STANDARDS

1. The authority citation for part 50 continues to read as follows:

Authority: 42 U.S.C. 7401, et seq.

2. Section 50.5 is amended by revising paragraphs (b) and (c) and by adding paragraphs (d) and (e) to read as follows:

§50.5 National secondary ambient air quality standards for sulfur oxides (sulfur dioxide).

* * * * *

(b) The level of the national secondary 1-hour ambient air quality standard for oxides of sulfur is 75 parts per billion (ppb, which is 1 part in 1,000,000,000), measured in the ambient air as sulfur dioxide (SO_2).

(c) The levels of the standards shall be measured by a reference method based on Appendix A-1 or A-2 of this part, or by a Federal Equivalent Method (FEM) designated in accordance with part 53 of this chapter.

(d) To demonstrate attainment with the 3-hour secondary standard, the second-highest 3-hour average must be based upon hourly data that are at least 75 percent complete in each calendar

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quarter. A 3-hour block average shall be considered valid only if all three hourly averages for the 3-hour period are available. If only one or two hourly averages are available, but the 3-hour average would exceed the level of the standard when zeros are substituted for the missing values, subject to the rounding rule of paragraph (a) of this section, then this shall be considered a valid 3-hour average. In all cases, the 3-hour block average shall be computed as the sum of the hourly averages divided by 3.

(e) The 1-hour secondary standard is met at an ambient air quality monitoring site when the three-year average of the annual 99th percentile of the daily maximum 1-hour average concentrations is less than or equal to 75 ppb, as determined in accordance with Appendix T of this part.

3. Section 50.11 is amended by deleting paragraphs (f) and (g) and revising paragraphs (a) through (e) to read as follows:

§50.11 National primary and secondary ambient air quality standards for oxides of nitrogen (with nitrogen dioxide as the indicator).

(a) The level of the national primary and secondary annual ambient air quality standards for oxides of nitrogen is 53 parts per billion (ppb, which is 1 part in 1,000,000,000), annual average concentration, measured in the ambient air as nitrogen dioxide.

(b) The level of the national primary and secondary 1-hour ambient air quality standards for oxides of nitrogen is 100 ppb, 1-hour average concentration, measured in the ambient air as nitrogen dioxide.

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(c) The levels of the standards shall be measured by:

(1) a reference method based on appendix F to this part; or

(2) a Federal equivalent method (FEM) designated in accordance with part 53 of this chapter.

(d) The annual primary and secondary standards are met when the annual average concentration in a calendar year is less than or equal to 53 ppb, as determined in accordance with Appendix S of this part for the annual standard.

(e) The 1-hour primary and secondary standards are met when the three-year average of the annual 98th percentile of the daily maximum 1-hour average concentration is less than or equal to 100 ppb, as determined in accordance with Appendix S of this part for the 1-hour standard.

4. Appendix S is amended as follows:

a. by revising paragraph 1.(a),

b. by revising the definition of "Design values" under paragraph 1.(c),

c. by revising paragraph 2.(b),

d. by revising paragraphs 3.1(a) through (d),

e. by revising paragraphs 3.2(a) through (e),

f. by revising paragraph 4.1(b),

g. by revising paragraph 4.2 (c),

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h. by revising paragraph 5.1(b), and

i. by revising paragraph 5.2(b) to read as follows:

Appendix S to Part 50—Interpretation of the Primary and Secondary National Ambient Air Quality Standards for Oxides of Nitrogen (Nitrogen Dioxide)

1. General.

(a) This appendix explains the data handling conventions and computations necessary for determining when the primary and secondary national ambient air quality standards for oxides of nitrogen as measured by nitrogen dioxide ("NO_2 NAAQS") specified in §50.11 are met. Nitrogen dioxide (NO_2) is measured in the ambient air by a Federal reference method (FRM) based on appendix F to this part or by a Federal equivalent method (FEM) designated in accordance with part 53 of this chapter. Data handling and computation procedures to be used in making comparisons between reported NO_2 concentrations and the levels of the NO_2 NAAQS are specified in the following sections.

* * * * *

(c) The terms used in this appendix are defined as follows:

* * * * *

Design values are the metrics (i.e., statistics) that are compared to the NAAQS levels to determine compliance, calculated as specified in section 5 of this appendix. The design values for the primary and secondary NAAQS are:

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(1) The annual mean value for a monitoring site for one year (referred to as the "annual primary or secondary standard design value").

(2) The 3-year average of annual 98th percentile daily maximum 1-hour values for a monitoring site (referred to as the "1-hour primary or secondary standard design value").

* * * * *

2. Requirements for Data Used for Comparisons with the NO_2 NAAQS and Data Reporting Considerations.

* * * * *

(b) When two or more NO_2 monitors are operated at a site, the state may in advance designate one of them as the primary monitor. If the state has not made this designation, the Administrator will make the designation, either in advance or retrospectively. Design values will be developed using only the data from the primary monitor, if this results in a valid design value. If data from the primary monitor do not allow the development of a valid design value, data solely from the other monitor(s) will be used in turn to develop a valid design value, if this results in a valid design value. If there are three or more monitors, the order for such comparison of the other monitors will be determined by the Administrator. The Administrator may combine data from different monitors in different years for the purpose of developing a valid 1-hour primary or secondary standard design value, if a valid design value cannot be developed solely with the data from a single monitor. However, data from two or more monitors in the same year at the same site will not be combined in an attempt to meet data completeness requirements, except if one monitor has physically replaced another instrument permanently, in which case the two

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instruments will be considered to be the same monitor, or if the state has switched the designation of the primary monitor from one instrument to another during the year.

* * * * *

3. Comparisons with the NO_2 NAAQS.

3.1 The Annual Primary and Secondary NO_2 NAAQS.

(a) The annual primary and secondary NO_2 NAAQS are met at a site when the valid annual primary standard design value is less than or equal to 53 parts per billion (ppb).

(b) An annual primary or secondary standard design value is valid when at least 75 percent of the hours in the year are reported.

(c) An annual primary or secondary standard design value based on data that do not meet the completeness criteria stated in section 3.1(b) may also be considered valid with the approval of, or at the initiative of, the Administrator, who may consider factors such as monitoring site closures/moves, monitoring diligence, the consistency and levels of the valid concentration measurements that are available, and nearby concentrations in determining whether to use such data.

(d) The procedures for calculating the annual primary and secondary standard design values are given in section 5.1 of this appendix.

3.2 The 1-hour Primary and Secondary NO_2 NAAQS.

(a) The 1-hour primary or secondary NO_2 NAAQS is met at a site when the valid 1-hour primary or secondary standard design value is less than or equal to 100 parts per billion (ppb).

(b) An NO_2 1-hour primary or secondary standard design value is valid if it encompasses three consecutive calendar years of complete data. A year meets data completeness requirements

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when all 4 quarters are complete. A quarter is complete when at least 75 percent of the sampling days for each quarter have complete data. A sampling day has complete data if 75 percent of the hourly concentration values, including state-flagged data affected by exceptional events which have been approved for exclusion by the Administrator, are reported.

(c) In the case of one, two, or three years that do not meet the completeness requirements of section 3.2(b) of this appendix and thus would normally not be useable for the calculation of a valid 3-year 1-hour primary or secondary standard design value, the 3-year 1-hour primary or secondary standard design value shall nevertheless be considered valid if one of the following conditions is true.

(i) At least 75 percent of the days in each quarter of each of three consecutive years have at least one reported hourly value, and the design value calculated according to the procedures specified in section 5.2 is above the level of the primary or secondary 1-hour standard.

(ii) (A) A 1-hour primary or secondary standard design value that is below the level of the NAAQS can be validated if the substitution test in section 3.2(c)(ii)(B) results in a "test design value" that is below the level of the NAAQS. The test substitutes actual ''high'' reported daily maximum 1-hour values from the same site at about the same time of the year (specifically, in the same calendar quarter) for unknown values that were not successfully measured. Note that the test is merely diagnostic in nature, intended to confirm that there is a very high likelihood that the original design value (the one with less than 75 percent data capture of hours by day and of days by quarter) reflects the true under-NAAQS-level status for that 3-year period; the result of this data substitution test (the ''test design value", as defined in section 3.2(c)(ii)(B)) is not considered the actual design value. For this test, substitution is permitted only if there are at least 200 days across the three matching quarters of the three years under consideration (which is

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about 75 percent of all possible daily values in those three quarters) for which 75 percent of the hours in the day, including state-flagged data affected by exceptional events which have been approved for exclusion by the Administrator, have reported concentrations. However, maximum 1-hour values from days with less than 75 percent of the hours reported shall also be considered in identifying the high value to be used for substitution.

(B) The substitution test is as follows: Data substitution will be performed in all quarter periods that have less than 75 percent data capture but at least 50 percent data capture, including state-flagged data affected by exceptional events which have been approved for exclusion by the Administrator; if any quarter has less than 50 percent data capture then this substitution test cannot be used. Identify for each quarter (e.g., January-March) the highest reported daily maximum 1-hour value for that quarter, excluding state-flagged data affected by exceptional events which have been approved for exclusion by the Administrator, looking across those three months of all three years under consideration. All daily maximum 1-hour values from all days in the quarter period shall be considered when identifying this highest value, including days with less than 75 percent data capture. If after substituting the highest non-excluded reported daily maximum 1-hour value for a quarter for as much of the missing daily data in the matching deficient quarter(s) as is needed to make them 100 percent complete, the procedure in section 5.2 yields a recalculated 3-year 1-hour standard "test design value" below the level of the standard, then the 1-hour primary or secondary standard design value is deemed to have passed the diagnostic test and is valid, and the level of the standard is deemed to have been met in that 3-year period. As noted in section 3.2(c)(i), in such a case, the 3-year design value based on the data actually reported, not the ''test design value'', shall be used as the valid design value.

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(iii) (A) A 1-hour primary or secondary standard design value that is above the level of the NAAQS can be validated if the substitution test in section 3.2(c)(iii)(B) results in a "test design value" that is above the level of the NAAQS. The test substitutes actual ''low" reported daily maximum 1-hour values from the same site at about the same time of the year (specifically, in the same three months of the calendar) for unknown values that were not successfully measured. Note that the test is merely diagnostic in nature, intended to confirm that there is a very high likelihood that the original design value (the one with less than 75 percent data capture of hours by day and of days by quarter) reflects the true above-NAAQS-level status for that 3-year period; the result of this data substitution test (the ''test design value", as defined in section 3.2(c)(iii)(B)) is not considered the actual design value. For this test, substitution is permitted only if there are a minimum number of available daily data points from which to identify the low quarter-specific daily maximum 1-hour values, specifically if there are at least 200 days across the three matching quarters of the three years under consideration (which is about 75 percent of all possible daily values in those three quarters) for which 75 percent of the hours in the day have reported concentrations. Only days with at least 75 percent of the hours reported shall be considered in identifying the low value to be used for substitution.

(B) The substitution test is as follows: Data substitution will be performed in all quarter periods that have less than 75 percent data capture. Identify for each quarter (e.g., January-March) the lowest reported daily maximum 1-hour value for that quarter, looking across those three months of all three years under consideration. All daily maximum 1-hour values from all days with at least 75 percent capture in the quarter period shall be considered when identifying this lowest value. If after substituting the lowest reported daily maximum 1-hour value for a quarter for as much of the missing daily data in the matching deficient quarter(s) as is needed to make them 75

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percent complete, the procedure in section 5.2 yields a recalculated 3-year 1-hour standard "test design value" above the level of the standard, then the 1-hour primary or secondary standard design value is deemed to have passed the diagnostic test and is valid, and the level of the standard is deemed to have been exceeded in that 3-year period. As noted in section 3.2(c)(i), in such a case, the 3-year design value based on the data actually reported, not the ''test design value'', shall be used as the valid design value.

(d) A 1-hour primary or secondary standard design value based on data that do not meet the completeness criteria stated in 3.2(b) and also do not satisfy section 3.2(c), may also be considered valid with the approval of, or at the initiative of, the Administrator, who may consider factors such as monitoring site closures/moves, monitoring diligence, the consistency and levels of the valid concentration measurements that are available, and nearby concentrations in determining whether to use such data.

(e) The procedures for calculating the 1-hour primary and secondary standard design values are given in section 5.2 of this appendix.

4. Rounding Conventions.

4.1 Rounding Conventions for the Annual Primary and Secondary NO_2 NAAQS.

* * * * *

(b) The annual primary or secondary standard design value is calculated pursuant to section 5.1 and then rounded to the nearest whole number or 1 ppb (decimals 0.5 and greater are rounded up to the nearest whole number, and any decimal lower than 0.5 is rounded down to the nearest whole number).

4.2 Rounding Conventions for the 1-hour Primary and Secondary NO_2 NAAQS.

* * * * *

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(c) The 1-hour primary or secondary standard design value is calculated pursuant to section 5.2 and then rounded to the nearest whole number or 1 ppb (decimals 0.5 and greater are rounded up to the nearest whole number, and any decimal lower than 0.5 is rounded down to the nearest whole number).

5. Calculation Procedures for the Primary and Secondary NO_2 NAAQS.

5.1 Procedures for the Annual Primary and Secondary NO_2 NAAQS

* * * * *

(b) The annual primary or secondary standard design value for a site is the valid annual mean rounded according to the conventions in section 4.1.

5.2 Calculation Procedures for the 1-hour Primary and Secondary NO_2 NAAQS.

* * * * *

(b) The 1-hour primary or secondary standard design value for a site is the mean of the three annual 98th percentile values, rounded according to the conventions in section 4.

* * * * *

5. Appendix T is amended as follows:

a. by revising paragraph 1.(a),

b. by revising the definition of "Design values" under paragraph 1.(c),

c. by revising paragraph 2.(b),

d. by revising paragraphs 3.(a) through (e),

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e. by revising paragraph 4.(c), and

f. by revising paragraph 5.(b) to read as follows:

Appendix T to Part 50—Interpretation of the Primary and Secondary National Ambient Air Quality Standards for Oxides of Sulfur (Sulfur Dioxide)

1. General.

(a) This appendix explains the data handling conventions and computations necessary for determining when the primary and secondary national ambient air quality standards for Oxides of Sulfur as measured by Sulfur Dioxide ("SO_2 NAAQS") specified in § 50.17 and §50.5 (b), respectively, are met at an ambient air quality monitoring site. Sulfur Dioxide (SO_2) is measured in the ambient air by a Federal reference method (FRM) based on appendix A-1 or A-2 to this part or by a Federal equivalent method (FEM) designated in accordance with part 53 of this chapter. Data handling and computation procedures to be used in making comparisons between reported SO_2 concentrations and the levels of the SO_2 NAAQS are specified in the following sections.

* * * * *

(c) The terms used in this appendix are defined as follows:

* * * * *

Design values are the metrics (i.e., statistics) that are compared to the NAAQS levels to determine compliance, calculated as specified in section 5 of this appendix. The design value for the primary and secondary 1-hour NAAQS is the 3-year average of annual 99th percentile daily

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maximum 1-hour values for a monitoring site (referred to as the "1-hour primary standard design value").

* * * * *

2. Requirements for Data Used for Comparisons with the SO_2 NAAQS and Data Reporting Considerations.

* * * * *

(b) Data from two or more monitors from the same year at the same site reported to EPA under distinct Pollutant Occurrence Codes shall not be combined in an attempt to meet data completeness requirements. The Administrator will combine annual 99th percentile daily maximum concentration values from different monitors in different years, selected as described here, for the purpose of developing a valid 1-hour primary or secondary standard design value. If more than one of the monitors meets the completeness requirement for all four quarters of a year, the steps specified in section 5(a) of this appendix shall be applied to the data from the monitor with the highest average of the four quarterly completeness values to derive a valid annual 99th percentile daily maximum concentration. If no monitor is complete for all four quarters in a year, the steps specified in section 3(c) and 5(a) of this appendix shall be applied to the data from the monitor with the highest average of the four quarterly completeness values in an attempt to derive a valid annual 99th percentile daily maximum concentration. This paragraph does not prohibit a monitoring agency from making a local designation of one physical monitor as the primary monitor for a Pollutant Occurrence Code and substituting the 1-hour data from a second physical monitor whenever a valid concentration value is not obtained from the primary monitor; if a monitoring agency substitutes data in this manner, each substituted value must be

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accompanied by an AQS qualifier code indicating that substitution with a value from a second physical monitor has taken place.

* * * * *

3. Comparisons with the 1-hour Primary and Secondary SO_2 NAAQS.

(a) The 1-hour primary or secondary SO_2 NAAQS is met at an ambient air quality monitoring site when the valid 1-hour primary or secondary standard design value is less than or equal to 75 parts per billion (ppb).

(b) An SO_2 1-hour primary or secondary standard design value is valid if it encompasses three consecutive calendar years of complete data. A year meets data completeness requirements when all 4 quarters are complete. A quarter is complete when at least 75 percent of the sampling days for each quarter have complete data. A sampling day has complete data if 75 percent of the hourly concentration values, including State-flagged data affected by exceptional events which have been approved for exclusion by the Administrator, are reported.

(c) In the case of one, two, or three years that do not meet the completeness requirements of section 3(b) of this appendix and thus would normally not be useable for the calculation of a valid 3-year 1-hour primary or secondary standard design value, the 3-year 1-hour primary or secondary standard design value shall nevertheless be considered valid if one of the following conditions is true.

(i) At least 75 percent of the days in each quarter of each of three consecutive years have at least one reported hourly value, and the design value calculated according to the procedures specified in section 5 is above the level of the primary or secondary 1-hour standard.

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(ii) (A) A 1-hour primary or secondary standard design value that is equal to or below the level of the NAAQS can be validated if the substitution test in section 3(c)(ii)(B) results in a "test design value" that is below the level of the NAAQS. The test substitutes actual ''high'' reported daily maximum 1-hour values from the same site at about the same time of the year (specifically, in the same calendar quarter) for unknown values that were not successfully measured. Note that the test is merely diagnostic in nature, intended to confirm that there is a very high likelihood that the original design value (the one with less than 75 percent data capture of hours by day and of days by quarter) reflects the true under-NAAQS-level status for that 3-year period; the result of this data substitution test (the ''test design value", as defined in section 3(c)(ii)(B)) is not considered the actual design value. For this test, substitution is permitted only if there are at least 200 days across the three matching quarters of the three years under consideration (which is about 75 percent of all possible daily values in those three quarters) for which 75 percent of the hours in the day, including State-flagged data affected by exceptional events which have been approved for exclusion by the Administrator, have reported concentrations. However, maximum 1-hour values from days with less than 75 percent of the hours reported shall also be considered in identifying the high value to be used for substitution.

(B) The substitution test is as follows: Data substitution will be performed in all quarter periods that have less than 75 percent data capture but at least 50 percent data capture, including State-flagged data affected by exceptional events which have been approved for exclusion by the Administrator; if any quarter has less than 50 percent data capture then this substitution test cannot be used. Identify for each quarter (e.g., January-March) the highest reported daily maximum 1-hour value for that quarter, excluding State-flagged data affected by exceptional events which have been approved for exclusion by the Administrator, looking across those three

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months of all three years under consideration. All daily maximum 1-hour values from all days in the quarter period shall be considered when identifying this highest value, including days with less than 75 percent data capture. If after substituting the highest reported daily maximum 1-hour value for a quarter for as much of the missing daily data in the matching deficient quarter(s) as is needed to make them 100 percent complete, the procedure in section 5 yields a recalculated 3-year 1-hour standard "test design value" less than or equal to the level of the standard, then the 1-hour primary or secondary standard design value is deemed to have passed the diagnostic test and is valid, and the level of the standard is deemed to have been met in that 3-year period. As noted in section 3(c)(i), in such a case, the 3-year design value based on the data actually reported, not the ''test design value'', shall be used as the valid design value.

(iii) (A) A 1-hour primary or secondary standard design value that is above the level of the NAAQS can be validated if the substitution test in section 3(c)(iii)(B) results in a "test design value" that is above the level of the NAAQS. The test substitutes actual ''low" reported daily maximum 1-hour values from the same site at about the same time of the year (specifically, in the same three months of the calendar) for unknown hourly values that were not successfully measured. Note that the test is merely diagnostic in nature, intended to confirm that there is a very high likelihood that the original design value (the one with less than 75 percent data capture of hours by day and of days by quarter) reflects the true above-NAAQS-level status for that 3-year period; the result of this data substitution test (the ''test design value", as defined in section 3(c)(iii)(B)) is not considered the actual design value. For this test, substitution is permitted only if there are a minimum number of available daily data points from which to identify the low quarter-specific daily maximum 1-hour values, specifically if there are at least 200 days across the three matching quarters of the three years under consideration (which is about 75 percent of

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all possible daily values in those three quarters) for which 75 percent of the hours in the day have reported concentrations. Only days with at least 75 percent of the hours reported shall be considered in identifying the low value to be used for substitution.

(B) The substitution test is as follows: Data substitution will be performed in all quarter periods that have less than 75 percent data capture. Identify for each quarter (e.g., January-March) the lowest reported daily maximum 1-hour value for that quarter, looking across those three months of all three years under consideration. All daily maximum 1-hour values from all days with at least 75 percent capture in the quarter period shall be considered when identifying this lowest value. If after substituting the lowest reported daily maximum 1-hour value for a quarter for as much of the missing daily data in the matching deficient quarter(s) as is needed to make them 75 percent complete, the procedure in section 5 yields a recalculated 3-year 1-hour standard "test design value" above the level of the standard, then the 1-hour primary or secondary standard design value is deemed to have passed the diagnostic test and is valid, and the level of the standard is deemed to have been exceeded in that 3-year period. As noted in section 3(c)(i), in such a case, the 3-year design value based on the data actually reported, not the ''test design value'', shall be used as the valid design value.

(d) A 1-hour primary or secondary standard design value based on data that do not meet the completeness criteria stated in 3(b) and also do not satisfy section 3(c), may also be considered valid with the approval of, or at the initiative of, the Administrator, who may consider factors such as monitoring site closures/moves, monitoring diligence, the consistency and levels of the valid concentration measurements that are available, and nearby concentrations in determining whether to use such data.

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(e) The procedures for calculating the 1-hour primary or secondary standard design values are given in section 5 of this appendix.

4. Rounding Conventions for the 1-hour Primary and Secondary SO_2 NAAQS.

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(c) The 1-hour primary or secondary standard design value is calculated pursuant to section 5 and then rounded to the nearest whole number or 1 ppb (decimals 0.5 and greater are rounded up to the nearest whole number, and any decimal lower than 0.5 is rounded down to the nearest whole number).

5. Calculation Procedures for the 1-hour Primary and Secondary SO_2 NAAQS.

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(b) The 1-hour primary or secondary standard design value for an ambient air quality monitoring site is the mean of the three annual 99th percentile values, rounded according to the conventions in section 4.

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