



Risk and Exposure Assessment for Review of the Secondary National Ambient Air Quality Standards for Oxides of Nitrogen and Oxides of Sulfur

Second Draft:

Main Content

EPA-452/P-09-004a
June 5, 2009

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

SECOND DRAFT:

MAIN CONTENT

U.S. Environmental Protection Agency
Office of Air Quality Planning and Standards
Research Triangle Park, NC

DISCLAIMER

This draft document has been prepared by staff from the Health and Environmental Impacts and Air Quality Analysis Divisions of the Office of Air Quality Planning and Standards, the Clean Air Markets Division, Office of Air Programs, the National Center for Environmental Assessment, Office of Research and Development, and the National Health and Environmental Effects Research Laboratory, Office of Research and Development, U.S. Environmental Protection Agency. Any opinions, findings, conclusions, or recommendations are those of the authors and do not necessarily reflect the views of EPA. This document is being circulated to obtain review and comment from the Clean Air Scientific Advisory Committee (CASAC) and the general public. Comments on this draft document should be addressed to Dr. Anne Rea, U.S. Environmental Protection Agency, Office of Air Quality Planning and Standards, C539-02, Research Triangle Park, North Carolina 27711 (email: rea.anne@epa.gov).

TABLE OF CONTENTS

List of Figures	iv
List of Tables	xiii
Acronyms and Abbreviations	xvii
Key Terms	xxi
Executive Summary	ES-1
1.0 Introduction	1-1
1.1 Rationale and Background for Joint Review	1-1
1.2 History	1-4
1.2.1 History of the Secondary NO ₂ NAAQS	1-4
1.2.2 History of the Secondary SO ₂ NAAQS	1-5
1.2.3 History of Related Assessments and Agency Actions	1-7
1.3 Scope of the Risk and Exposure Assessment for the Current Review	1-9
1.3.1 Species of Nitrogen Included in the Analyses	1-9
1.3.2 Species of Sulfur Included in the Analyses	1-11
1.3.3 Overview of Nitrogen- and Sulfur-Related Ecological Effects	1-12
1.4 Policy-Relevant Questions	1-16
1.5 References	1-21
2.0 Overview of Risk and Exposure Assessment	2-1
2.1 Introduction	2-1
2.2 Seven-Step Approach	2-7
2.3 Linkages for Structuring Ecologically Relevant Standards	2-9
2.4 Ecosystem Services	2-10
2.4.1 Aquatic Acidification	2-15
2.4.2 Terrestrial Acidification	2-15
2.4.3 Aquatic Nutrient Enrichment	2-15
2.4.4 Terrestrial Nutrient Enrichment	2-16
2.4.5 Sulfur and Mercury Methylation	2-16
2.5 Uncertainty	2-18
2.6 References	2-19
3.0 Sources, Ambient Concentrations, and Deposition	3-1
3.1 Science Overview	3-2
3.2 Nationwide Sources, Concentrations, and Deposition of NO _x , NH ₃ , and SO _x	3-3
3.2.1 Sources of Nitrogen and Sulfur	3-3
3.2.2 Nationwide Atmospheric Concentrations of NO _x and SO _x	3-10
3.2.3 Nationwide Deposition of Nitrogen and Sulfur	3-14
3.2.4 Policy-Relevant Background Concentrations	3-21
3.2.5 Nonambient Loadings of Nitrogen and Sulfur	3-22
3.3 Spatial and Temporal Characterization of Deposition for Case Study Areas	3-22
3.3.1 Purpose and Intent	3-22
3.3.2 Data and Analytical Techniques	3-23
3.3.3 Characterization of Deposition in Case Study Areas	3-25

3.4	Contributions of Emissions of NO _x and NH ₃ to Deposition of Nitrogen	3-80
3.4.1	Purpose and Intent.....	3-80
3.4.2	Analytical Techniques	3-80
3.4.3	Results and Findings.....	3-81
3.4.4	Summary of Findings.....	3-83
3.5	Discussion of Uncertainties	3-90
3.6	References.....	3-96
4.0	Acidification	4-1
4.1	Science Overview	4-1
4.1.1	Aquatic Acidification.....	4-2
4.1.2	Terrestrial Acidification.....	4-3
4.2	Aquatic Acidification.....	4-3
4.2.1	Ecological Indicators, Ecological Responses, and Ecosystem Services.....	4-5
4.2.2	Characteristics of Sensitive Areas	4-8
4.2.3	Case Study Area Selection.....	4-10
4.2.4	Current Conditions in Case Study Areas	4-13
4.2.5	Degree of Extrapolation to Larger Assessment Areas.....	4-31
4.2.6	Current Conditions for the Adirondack Case Study Area and the Shenandoah Case Study Area.....	4-34
4.2.7	Ecological Effect Function for Aquatic Acidification.....	4-36
4.2.8	Uncertainty and Variability.....	4-39
4.3	Terrestrial Acidification.....	4-41
4.3.1	Ecological Indicators, Ecological Responses, and Ecosystem Services.....	4-41
4.3.2	Characteristics of Sensitive Areas	4-50
4.3.3	Case Study Selection.....	4-51
4.3.4	Current Conditions Assessment.....	4-53
4.3.5	Results for the Case Study Areas.....	4-57
4.3.6	Evaluation of Representativeness of Case Study Areas	4-61
4.3.7	Current Conditions for Sugar Maple and Red Spruce	4-63
4.3.8	Ecological Effect Function for Terrestrial Acidification.....	4-66
4.3.9	Uncertainty and Variability.....	4-69
4.4	References.....	4-72
5.0	Nutrient Enrichment.....	5-1
5.1	Science Overview	5-1
5.1.1	Aquatic Nutrient Enrichment.....	5-2
5.1.2	Terrestrial Nutrient Enrichment.....	5-3
5.2	Aquatic Nutrient Enrichment.....	5-3
5.2.1	Ecological Indicators, Ecological Responses, and Ecosystem Services.....	5-5
5.2.2	Characteristics of Sensitive Areas	5-15
5.2.3	Case Study Selection.....	5-16
5.2.4	Current Conditions in the Case Study Areas	5-18
5.2.5	Degree of Extrapolation to Larger Assessment Areas.....	5-28
5.2.6	Current Conditions for Other/Additional Estuaries	5-31
5.2.7	Ecological Effect Function for Aquatic Nutrient Enrichment.....	5-32
5.2.8	Uncertainty and Variability.....	5-40

5.3	Terrestrial Nutrient Enrichment.....	5-43
5.3.1	Ecological Indicators, Ecological Responses, and Ecosystem Services.....	5-44
5.3.2	Characteristics of Sensitive Areas	5-54
5.3.3	Case Study Selection.....	5-56
5.3.4	Current Conditions in Case Study Areas	5-58
5.3.5	Degree of Extrapolation to Larger Assessment Areas.....	5-70
5.3.6	Current Conditions for Select Locations Nationwide.....	5-73
5.3.7	Ecological Effect Function for Terrestrial Nutrient Enrichment.....	5-81
5.3.8	Uncertainty and Variability.....	5-81
5.4	Conclusions.....	5-83
5.5	References.....	5-83
6.0	Additional Effects	6-1
6.1	Visibility, Climate, and Materials.....	6-1
6.2	Sulfur and Mercury Methylation	6-2
6.2.1	Science Background.....	6-2
6.2.2	Qualitative Analysis.....	6-4
6.3	Nitrous Oxide.....	6-11
6.4	Nitrogen Addition Effects on Primary Productivity and Biogenic Greenhouse Gas Fluxes.....	6-13
6.4.1	Effects on Primary Productivity and Carbon Budgeting.....	6-13
6.4.2	Biogenic Emissions of Nitrous Oxide.....	6-19
6.4.3	Methane Emissions and Uptake.....	6-20
6.4.4	Emission Factors.....	6-22
6.4.5	Uncertainty.....	6-23
6.5	Direct Phytotoxic Effects of Gaseous SO _x AND NO _x	6-24
6.5.1	SO ₂	6-24
6.5.2	NO, NO ₂ and Peroxyacetyl Nitrate (PAN)	6-25
6.5.3	Nitric Acid (HNO ₃).....	6-27
6.6	References.....	6-29
7.0	Synthesis and Integration of Case Study Results.....	7-1
7.1	Summary of Ecological Responses, Ecological Effects, and Welfare Effects	7-5
7.1.1	Ecological Responses.....	7-5
7.1.2	Ecological Effects	7-6
7.1.3	Ecological Benefits/Welfare Effects.....	7-17
7.2	Uncertainty.....	7-21
7.3	Conclusions.....	7-24
7.4	References.....	7-28

APPENDICES

Appendix 1	Description of CMAQ Applications and Model Performance Evaluation
Appendix 2	Trends in Wet Deposition at NADP Sites
Appendix 3	Components of Reactive Nitrogen Deposition: 2002–2005

Appendix 4	Aquatic Acidification Case Study
	Attachment A Modeling Descriptions
	Attachment B EMAP/TIME/LTM Programs
Appendix 5	Terrestrial Acidification Case Study
	Attachment A Relationship Between Atmospheric Nitrogen and Sulfur Deposition and Sugar Maple and Red Spruce Tree Growth
Appendix 6	Aquatic Nutrient Enrichment Case Study
Appendix 7	Terrestrial Nutrient Enrichment Case Study
Appendix 8	Analysis of Ecosystem Services Impacts for the NO _x /SO _x Secondary NAAQS Review

LIST OF FIGURES

Figure 1.3-1.	Schematic diagram of the cycle of reactive, oxidized nitrogen species in the atmosphere. Particulate-phase organic nitrates are also formed from the species on the right side of the figure (U.S. EPA, 2008).....	1-11
Figure 1.3-2.	Schematic diagram of the cycle of sulfur species in the atmosphere.....	1-12
Figure 1.3-3.	Nitrogen and sulfur cycling and interactions in the environment.	1-16
Figure 1.4-1.	Possible structure of a secondary NAAQS for NO _x and SO _x based on an ecological indicator.	1-20
Figure 2.1-1.	National map highlighting the 8 case study areas and the Rocky Mountain National Park (a supplemental study area) evaluated in the Risk and Exposure Assessment.....	2-5
Figure 2.3-1.	Possible structure of a secondary NAAQS for NO _x and SO _x based on an ecological indicator.	2-9
Figure 2.4-1.	Millennium ecosystem assessment categorization of ecosystem services and their links to human well-being (MEA, 2005a).....	2-11
Figure 2.4-2.	Pathway from nitrogen deposition to valuation for an aquatic system.	2-14
Figure 3.2-1.	Annual NO _x emissions across major source categories in 2002.	3-5
Figure 3.2-2.	Spatial distribution of annual total NO _x emissions (tons/yr) for 2002.	3-6
Figure 3.2-3.	Spatial distribution of annual total NH ₃ emissions (tons/yr) for 2002.	3-7
Figure 3.2-4.	Annual SO ₂ emissions across major source categories in 2002.	3-9
Figure 3.2-5.	Spatial distribution of annual total SO ₂ emissions (tons/yr) for 2002.....	3-10
Figure 3.2-6.	Model-predicted annual average NO _y concentrations (ppb) for 2002.....	3-12
Figure 3.2-7.	Model-predicted annual average SO ₂ concentrations (ppb) for 2002.	3-14
Figure 3.2-8.	Total wet plus dry oxidized nitrogen deposition (kg N/ha/yr) in 2002.	3-18
Figure 3.2-9.	Total wet plus dry reduced nitrogen deposition (kg N/ha/yr) in 2002.	3-19
Figure 3.2-10.	Total reactive nitrogen deposition (kg N/ha/yr) in 2002.	3-20
Figure 3.2-11.	Total wet and dry sulfur deposition (kg S/ha/yr) in 2002.	3-21
Figure 3.3-1a.	Annual total reactive nitrogen deposition (kg N/ha/yr) from 2002 through 2005 for each case study area in the East.....	3-29
Figure 3.3-1b.	Annual total reactive nitrogen deposition (kg N/ha/yr) from 2002 through 2005 for case study areas in the West, and the Rocky Mountain National Park.....	3-29

Figure 3.3-2.	Relative amounts of oxidized and reduced nitrogen deposition in 2002 for case study areas and the Rocky Mountain National Park.	3-31
Figure 3.3-3a.	Components of total reactive nitrogen deposition for 2002 in the Adirondack Case Study Area.	3-32
Figure 3.3-3b.	Components of total reactive nitrogen deposition for 2002 in the Hubbard Brook Experimental Forest Case Study Area.	3-32
Figure 3.3-3c.	Components of total reactive nitrogen deposition for 2002 in the Kane Experimental Forest Case Study Area.	3-33
Figure 3.3-3d.	Components of total reactive nitrogen deposition for 2002 in the Neuse River/Neuse River Estuary Case Study Area.	3-33
Figure 3.3-3e.	Components of total reactive nitrogen deposition for 2002 in the Potomac River/Potomac Estuary Case Study Area.	3-34
Figure 3.3-3f.	Components of total reactive nitrogen deposition for 2002 in the Shenandoah Case Study Area.	3-34
Figure 3.3-3g.	Components of total reactive nitrogen deposition for 2002 in the Rocky Mountain National Park.	3-35
Figure 3.3-3h.	Components of total reactive nitrogen deposition for 2002 in the Sierra Nevada Range portion of the Mixed Conifer Forest Case Study Area.	3-35
Figure 3.3-3i.	Components of total reactive nitrogen deposition for 2002 in the Transverse Range portion of the Mixed Conifer Forest Case Study Area.	3-36
Figure 3.3-4a.	Annual total dry plus wet reactive nitrogen deposition (kg N/ha/yr) in 2002 for the case study areas in the East.	3-40
Figure 3.3-4b.	Annual total dry plus wet oxidized nitrogen deposition (kg N/ha/yr) in 2002 for the case study areas in the East.	3-41
Figure 3.3-4c.	Annual total dry plus wet reduced nitrogen deposition (kg N/ha/yr) in 2002 for the case study areas in the East.	3-42
Figure 3.3-4d.	Annual total wet reactive nitrogen deposition (kg N/ha/yr) in 2002 for the case study areas in the East.	3-43
Figure 3.3-4e.	Annual total dry reactive nitrogen deposition (kg N/ha/yr) in 2002 for the case study areas in the East.	3-44
Figure 3.3-5a.	Annual total dry plus wet reactive nitrogen deposition (kg N/ha/yr) in 2002 for case study areas and Rocky Mountain National Park in the West.	3-45
Figure 3.3-5b.	Annual total dry plus wet oxidized nitrogen deposition (kg N/ha/yr) in 2002 for case study areas and Rocky Mountain National Park in the West.	3-46
Figure 3.3-5c.	Annual total dry plus wet reduced nitrogen deposition (kg N/ha/yr) in 2002 for case study areas and Rocky Mountain National Park in the West.	3-47
Figure 3.3-6a.	Percentage of 2002 total reactive nitrogen deposition in the Adirondack Case Study Area.	3-49
Figure 3.3-6b.	Percentage of 2002 total reactive nitrogen deposition in the Hubbard Brook Experimental Forest Case Study Area.	3-49
Figure 3.3-6c.	Percentage of 2002 total reactive nitrogen deposition in the Kane Experimental Forest Case Study Area.	3-50

Figure 3.3-6d.	Percentage of 2002 total reactive nitrogen deposition in the Potomac River/Potomac Estuary Case Study Area.	3-50
Figure 3.3-6e.	Percentage of 2002 total reactive nitrogen deposition in the Shenandoah Case Study Area.	3-51
Figure 3.3-6f.	Percentage of 2002 total reactive nitrogen deposition in the Neuse River/Neuse River Estuary Case Study Area.	3-51
Figure 3.3-6g.	Percentage of 2002 total reactive nitrogen deposition in the Rocky Mountain National Park.	3-52
Figure 3.3-6h.	Percentage of 2002 total reactive nitrogen deposition in the Sierra Nevada Range portion of the Mixed Conifer Forest Case Study Area.	3-52
Figure 3.3-6i.	Percentage of 2002 total reactive nitrogen deposition in the Transverse Range portion of the Mixed Conifer Forest Case Study Area.	3-53
Figure 3.3-7a.	Percentage of 2002 reactive nitrogen deposition for each component of nitrogen deposition in the Adirondack Case Study Area.	3-53
Figure 3.3-7b.	Percentage of 2002 reactive nitrogen deposition for each component of nitrogen deposition in the Hubbard Brook Experimental Forest Case Study Area.	3-54
Figure 3.3-7c.	Percentage of 2002 reactive nitrogen deposition for each component of nitrogen deposition in the Kane Experimental Forest Case Study Area.	3-54
Figure 3.3-7d.	Percentage of 2002 reactive nitrogen deposition for each component of nitrogen deposition in the Potomac River/Potomac Estuary Case Study Area.	3-55
Figure 3.3-7e.	Percentage of 2002 reactive nitrogen deposition for each component of nitrogen deposition in the Shenandoah Case Study Area.	3-55
Figure 3.3-7f.	Percentage of 2002 reactive nitrogen deposition for each component of nitrogen deposition in the Neuse River/Neuse River Estuary Case Study Area.	3-56
Figure 3.3-7g.	Percentage of 2002 reactive nitrogen deposition for each component of nitrogen deposition in the Rocky Mountain National Park.	3-56
Figure 3.3-7h.	Percentage of 2002 reactive nitrogen deposition for each component of nitrogen deposition in the Sierra Nevada Range portion of the Mixed Conifer Forest Case Study Area.	3-57
Figure 3.3-7i.	Percentage of 2002 reactive nitrogen deposition for each component of nitrogen deposition in the Transverse Range portion of the Mixed Conifer Forest Case Study Area.	3-57
Figure 3.3-8.	Percentage of 2002 NH ₃ emissions by season for each state containing a case study area.	3-58
Figure 3.3-9a.	Annual sulfur deposition (kg S/ha/yr) from 2002 through 2005 for each case study area in the East.	3-60
Figure 3.3-9b.	Annual sulfur deposition (kg S/ha/yr) from 2002 through 2005 for case study areas in the West, as well as the Rocky Mountain National Park.	3-60
Figure 3.3-10.	Relative amount of wet and dry annual sulfur deposition in 2002 for case study areas.	3-62
Figure 3.3-11.	Relative amount of wet and dry annual sulfur deposition based on deposition for the period 2002 through 2005 for each case study area and the Rocky Mountain National Park.	3-62

Figure 3.3-12a. Annual total dry plus wet sulfur deposition (kg S/ha/yr) in 2002 for the case study areas in the East.	3-65
Figure 3.3-12b. Annual wet sulfur deposition (kg S/ha/yr) in 2002 for the case study areas in the East.	3-66
Figure 3.3-12c. Annual dry sulfur deposition (kg S/ha/yr) in 2002 for the case study areas in the East.	3-67
Figure 3.3-13. Annual total dry plus wet sulfur deposition (kg S/ha/yr) in 2002 for case study areas and Rocky Mountain National Park in the West.	3-68
Figure 3.3-14a. Percentage of 2002 total sulfur deposition in the Adirondack Case Study Area.	3-70
Figure 3.3-14b. Percentage of 2002 total sulfur deposition in the Hubbard Brook Experimental Forest Case Study Area.	3-70
Figure 3.3-14c. Percentage of 2002 total sulfur deposition in the Kane Experimental Forest Case Study Area.	3-71
Figure 3.3-14d. Percentage of 2002 total sulfur deposition in the Potomac River/Potomac Estuary Case Study Area.	3-71
Figure 3.3-14e. Percentage of 2002 total sulfur deposition in the Shenandoah Case Study Area.	3-72
Figure 3.3-14f. Percentage of 2002 total sulfur deposition in the Neuse River/Neuse River Estuary Case Study Area.	3-72
Figure 3.3-14g. Percentage of 2002 total sulfur deposition in the Rocky Mountain National Park.	3-73
Figure 3.3-14h. Percentage of 2002 total sulfur deposition in the Sierra Nevada Range portion of the Mixed Conifer Forest Case Study Area.	3-73
Figure 3.3-14i. Percentage of 2002 total sulfur deposition in the Transverse Range portion of the Case Study Area.	3-74
Figure 3.3-15a. Percentage of 2002 deposition for each component of sulfur deposition in the Adirondack Case Study Area.	3-74
Figure 3.3-15b. Percentage of 2002 deposition for each component of sulfur deposition in the Hubbard Brook Experimental Forest Case Study Area.	3-75
Figure 3.3-15c. Percentage of 2002 deposition for each component of sulfur deposition in the Kane Experimental Forest Case Study Area.	3-75
Figure 3.3-15d. Percentage of 2002 deposition for each component of sulfur deposition in the Potomac River/Potomac Estuary Case Study Area.	3-76
Figure 3.3-15e. Percentage of 2002 deposition for each component of sulfur deposition in the Shenandoah Case Study Area.	3-76
Figure 3.3-15f. Percentage of 2002 deposition for each component of sulfur deposition in the Neuse River/Neuse River Estuary Case Study Area.	3-77
Figure 3.3-15g. Percentage of 2002 deposition for each component of sulfur deposition in the Rocky Mountain National Park.	3-77
Figure 3.3-15h. Percentage of 2002 deposition for each component of sulfur deposition in the Sierra Nevada Range portion of the Mixed Conifer Forest Case Study Area.	3-78
Figure 3.3-15i. Percentage of 2002 deposition for each component of sulfur deposition in the Transverse Range portion of the Mixed Conifer Forest Case Study Area.	3-78

Figure 3.4-1.	The percentage impacts of a 50% decrease in NO _x emissions on total reactive nitrogen deposition in the East.....	3-84
Figure 3.4-2.	The percentage impacts of a 50% decrease in NO _x emissions on oxidized nitrogen deposition in the East.	3-85
Figure 3.4-3.	The percentage impacts of a 50% decrease in NO _x emissions on reduced nitrogen deposition in the East.	3-86
Figure 3.4-4.	The percentage impacts of a 50% decrease in NH ₃ emissions on total reactive nitrogen deposition in the East.....	3-87
Figure 3.4-5.	The percentage impacts of a 50% decrease in NH ₃ emissions on oxidized nitrogen deposition in the East.	3-88
Figure 3.4-6.	The percentage impacts of a 50% decrease in NH ₃ emissions on reduced nitrogen deposition in the East.	3-89
Figure 3.4-7.	The percentage impacts of a 50% decrease in SO _x emissions on sulfur deposition in the East.....	3-90
Figure 3.5-1.	Fine-scale and 12-km annual total oxidized nitrogen deposition for the Adirondack Case Study Area and the surrounding region.	3-94
Figure 3.5-2.	Fine-scale and 12-km annual total reduced nitrogen deposition for the Adirondack Case Study Area and the surrounding region.	3-95
Figure 3.5-3.	Fine-scale and 12-km annual total sulfur deposition for the Adirondack Case Study Area and the surrounding region.	3-96
Figure 4.2-1.	(a) Number of fish species per lake or stream versus acidity, expressed as acid neutralizing capacity for Adirondack Case Study Area lakes (Sullivan et al., 2006). (b) Number of fish species among 13 streams in Shenandoah National Park. Values of acid neutralizing capacity are means based on quarterly measurements from 1987 to 1994. The regression analysis shows a highly significant relationship ($p < .0001$) between mean stream acid neutralizing capacity and the number of fish species.....	4-6
Figure 4.2-2.	Ecosystems sensitive to acidifying deposition in the eastern United States (U.S. EPA, modified from NAPAP, 2005).	4-10
Figure 4.2-3.	Annual average total wet deposition (kg/ha/yr) for the period 1990 to 2006 in SO ₄ ²⁻ (green) and NO ₃ ⁻ (blue) from eight NADP/NTN sites in the Adirondack Case Study Area.	4-12
Figure 4.2-4.	Air pollution concentrations and deposition for the period 1990 to 2006 using one CASTNET and seven NADP/NTN sites in the Shenandoah Case Study Area. (a) Annual average air concentrations of SO ₂ (blue), oxidized nitrogen (red), SO ₄ ²⁻ (green), and reduced nitrogen (black). (b) Annual average total wet deposition (kg/ha/yr) of SO ₄ ²⁻ (green) and NO ₃ ⁻ (blue).....	4-13
Figure 4.2-5.	(Top) The location of lakes in the Adirondack Case Study Area used for MAGIC (red dots) and critical load (green dots) modeling sites. (Bottom) The location of streams used for both MAGIC and critical load modeling for the Shenandoah Case Study Area.	4-15
Figure 4.2-6.	Trends over time for SO ₄ ²⁻ , NO ₃ ⁻ , and acid neutralizing capacity in LTM. SO ₄ ²⁻ and NO ₃ ⁻ concentrations have decreased in surface waters by approximately 26% and 13%, respectively.	4-18

Figure 4.2-7.	NO ₃ ⁻ concentrations of preacidification (1860) and current (2006) conditions based on hindcasts of 44 lakes in the Adirondack Case Study Area modeled using MAGIC.....	4-19
Figure 4.2-8.	SO ₄ ²⁻ concentrations of preacidification (1860) and current (2006) conditions based on hindcasts of 44 lakes in the Adirondack Case Study Area modeled using MAGIC.....	4-19
Figure 4.2-9.	Acid neutralizing capacity concentrations from 94 lakes in the Adirondack Case Study Area. Monitoring data from the TIME/LTM programs.	4-20
Figure 4.2-10.	Acid neutralizing capacity concentrations of preacidification (1860) and current (2006) conditions based on hindcasts of 44 modeled lakes in the Adirondack Case Study Area.	4-20
Figure 4.2-11.	Percentage of Adirondack Case Study Area lakes in the five classes of acidification (i.e., Acute, Severe, Elevated, Moderate, Low) for years 2006 and 1860 (preacidification) for 44 lakes modeled using MAGIC. Error bar indicates the 95% confidence interval.	4-21
Figure 4.2-12.	Critical loads of acidifying deposition that each surface waterbody in the Adirondack Case Study Area can receive while maintaining or exceeding an acid neutralizing capacity concentration of 50 µeq/L based on 2002 data. Watersheds with critical load values <100 meq/m ² /yr (red and orange dots) are most sensitive to surface water acidification, whereas watersheds with values >100 meq/m ² .yr (yellow and green dots) are the least sensitive sites.....	4-22
Figure 4.2-13.	Critical load exceedances (red dots) based on 2002 deposition magnitudes for Adirondack Case Study Area waterbodies where the critical limit acid neutralizing capacity is 0, 20, 50, and 100 µeq/L, respectively. Green dots represent lakes where current total nitrogen and sulfur deposition is below the critical load. See Table 4.2-3.....	4-23
Figure 4.2-14.	Trends over time for SO ₄ ²⁻ (blue), NO ₃ ⁻ (green) and acid neutralizing capacity (red) concentrations in VTSSS LTM-monitored streams in the Shenandoah Case Study Area.....	4-25
Figure 4.2-15.	NO ₃ ⁻ concentrations of preacidification (1860) and current (2006) conditions based on hindcasts of 60 streams modeled using MAGIC in the Shenandoah Case Study Area.....	4-26
Figure 4.2-16.	SO ₄ ²⁻ concentrations of preacidification (1860) and current (2006) conditions based on hindcasts of 60 streams modeled using MAGIC in the Shenandoah Case Study Area.....	4-27
Figure 4.2-17.	Acid neutralizing capacity concentrations from 68 streams in the VTSSS-SWAS/LTM monitoring network in the Shenandoah Case Study Area (2006 data).	4-27
Figure 4.2-18.	Acid neutralizing capacity concentrations of preacidification (1860) and current (2006) conditions based on hindcasts of 60 streams modeled using MAGIC in the Shenandoah Case Study Area.....	4-28
Figure 4.2-19.	Percentage of streams in the five classes of acidification (i.e., Acute, Severe, Elevated, Moderate, Low Concern) for years 2006 and 1860 (pre-acidification) for 60 streams modeled using MAGIC in the	

	Shenandoah Case Study Area. The number of streams in each class is above the bar. Error bars indicate the 95% confidence interval.	4-28
Figure 4.2-20.	Critical loads of surface water acidity for an acid neutralizing capacity concentration of 50 $\mu\text{eq/L}$ for streams in the Shenandoah Case Study Area. Each dot represents an estimated amount of acidifying deposition (i.e., critical load) that each stream's watershed can receive and still maintain a surface water acid neutralizing capacity concentration $>50 \mu\text{eq/L}$. Watersheds with critical load values $<100 \text{ meq/m}^2/\text{yr}$ (red and orange dots) are most sensitive to surface water acidification, whereas watersheds with values $>100 \text{ meq/m}^2/\text{yr}$ (yellow and green dots) are the least sensitive sites.	4-29
Figure 4.2-21.	Critical load exceedances for acid neutralizing capacity concentrations of 0, 20, 50, and 100 $\mu\text{eq/L}$ for streams in the Shenandoah Case Study Area. Green dots represent lakes where current total nitrogen and sulfur deposition is below the critical load and that maintain an acid neutralizing capacity concentration of 0, 20, 50, and 100 $\mu\text{eq/L}$, respectively. Red dots represent streams where current total nitrogen and sulfur deposition exceeds the critical load, indicating they are currently impacted by acidifying deposition. See Table 4.2-5.	4-30
Figure 4.2-22.	The depositional load function defined by the model.	4-38
Figure 4.2-23	Deposition load graphs for Clear Pond and Middle Flow Lake, New York.	4-39
Figure 4.3-1.	The relationship between the Bc/Al ratio in soil solution and the percentage of tree species (found growing in North America) exhibiting a 20% reduction in growth relative to controls (after Sverdrup and Warfvinge, 1993).	4-43
Figure 4.3-2.	2006 annual value of sugar maple and red spruce harvests and maple syrup production, by state.	4-48
Figure 4.3-3.	Map of areas of potential sensitivity of red spruce and sugar maple to acidification in the United States (see Table 1.2-1 of Appendix 5 for a listing of data sources to produce this map).	4-53
Figure 4.3-4.	The critical load function created from the calculated maximum and minimum levels of total nitrogen and sulfur deposition (eq/ha/yr). The grey areas show deposition levels less than the established critical loads. The red line is the maximum amount of total sulfur deposition (valid only when nitrogen deposition is less than the minimum critical level of nitrogen deposition [blue dotted line]) in the critical load. The flat line portion of the curves indicates nitrogen deposition corresponding to the $\text{CL}_{\text{min}}(\text{N})$ (nitrogen absorbed by nitrogen sinks within the system).	4-55
Figure 4.3-5.	Critical load function response curves for the three selected critical loads conditions (corresponding to the three levels of protection) for the Kane Experimental Forest Case Study Area. The 2002 CMAQ/NADP total nitrogen and sulfur (N+S_{comb}) deposition was greater than the highest and intermediate level of protection critical loads. The flat line portion of the curves indicates total nitrogen deposition corresponding to the $\text{CL}_{\text{min}}(\text{N})$ (nitrogen absorbed by nitrogen sinks within the system).	4-59

Figure 4.3-6.	Critical load function response curves for the three selected critical loads conditions (corresponding to the three levels of protection) for the Hubbard Brook Experimental Forest Case Study Area. The 2002 CMAQ/NADP total nitrogen and sulfur ($N+S_{\text{comb}}$) deposition was greater than the highest level of protection critical load. The flat line portion of the curves indicates total nitrogen deposition corresponding to the $CL_{\text{min}}(N)$ (nitrogen absorbed by nitrogen sinks within the system).	4-59
Figure 4.3-7.	The influence of the 2002 CMAQ/NADP total reduced nitrogen (NH_x-N) deposition on the critical function response curve, and in turn, the maximum amounts of sulfur ($CL_{\text{max}}(S)$) and oxidized nitrogen (NO_x-N) in the critical load for the Kane Experimental Forest Case Study Area. The critical load of oxidized nitrogen (NO_x-N) is 661 eq/ha/yr (910–249). The $CL_{\text{min}}(N)$ (nitrogen absorbed by nitrogen sinks within the system) corresponds to the value depicted in Figure 4.3-5.	4-60
Figure 4.3-8.	The influence of the 2002 CMAQ/NADP total reduced nitrogen (NH_x-N) deposition on the critical load function response curve and, in turn, the maximum amounts of sulfur ($CL_{\text{max}}(S)$) and oxidized nitrogen (NO_x-N) in the critical load for the Hubbard Brook Experimental Forest Case Study Area. The critical load of oxidized nitrogen (NO_x-N) is 328 eq/ha/yr (487–159). The $CL_{\text{min}}(N)$ (nitrogen absorbed by nitrogen sinks within the system) corresponds to the value depicted in Figure 4.3-6.	4-60
Figure 4.3.9.	The extreme (i.e., highest and lowest) and average critical load function response curves for the three levels of protection ($(Bc/Al)_{\text{crit}} = 0.6, 1.2,$ and 10.0) for the critical load assessments for the full range of sugar maple. The $CL_{\text{min}}(N)$ value for all curves is 42.86 eq/ha/yr, but it is not shown in the figure.	4-68
Figure 4.3.10.	The extreme (i.e., highest and lowest) and average critical load function response curves for the three levels of protection ($(Bc/Al)_{\text{crit}} = 0.6, 1.2,$ and 10.0) for the critical load assessments for the full range of red spruce. The $CL_{\text{min}}(N)$ value for all curves is 42.86 eq/ha/yr, but it is not shown in the figure.	4-69
Figure 5.2-1.	Descriptions of the five eutrophication indicators used in the NEEA (Bricker et al., 2007).....	5-6
Figure 5.2-2.	ASSETS EI response curve.....	5-9
Figure 5.2-3.	Areas potentially sensitive to aquatic nutrient enrichment.....	5-16
Figure 5.2-4.	Atmospheric deposition yields of oxidized nitrogen over the Potomac River and Potomac Estuary watershed.....	5-21
Figure 5.2-5.	Atmospheric deposition yields of reduced nitrogen over the Potomac River and Potomac Estuary watershed.....	5-21
Figure 5.2-6.	Atmospheric deposition yields of total nitrogen over the Potomac River and Potomac Estuary watershed.....	5-22
Figure 5.2-7.	Total nitrogen yields from all sources as predicted using version 3 of the Chesapeake Bay SPARROW application with updated 2002 atmospheric deposition inputs.....	5-23
Figure 5.2-8.	Atmospheric deposition yields of oxidized nitrogen over the Neuse River and Neuse River Estuary watershed.....	5-25

Figure 5.2-9.	Atmospheric deposition yields of reduced nitrogen over the Neuse River and Neuse River Estuary watershed.	5-25
Figure 5.2-10.	Atmospheric deposition yields of total nitrogen over the Neuse River and Neuse River Estuary watershed.	5-26
Figure 5.2-11.	Total nitrogen yields from all sources predicted by a SPARROW application for the Neuse, Tar-Pamlico, and Cape Fear watersheds with 2002 data inputs.	5-27
Figure 5.2-12.	Preliminary classifications of estuary typology across the nation (Bricker et al., 2007).	5-30
Figure 5.2-13.	ASSETS EI scores for 48 systems examined in the 2007 NEEA Update (Bricker et al., 2007).	5-32
Figure 5.2-14.	Response curve relating instream total nitrogen concentration (TNs) to total nitrogen atmospheric deposition load (TN _{atm}) for the Potomac River watershed.	5-34
Figure 5.2-15.	Example of fitted OEC curve for target ASSETS EI=2 for the Potomac Estuary.	5-34
Figure 5.2-16.	Response curve relating instream total nitrogen concentration to total nitrogen atmospheric deposition load for the Neuse River/Neuse River Estuary Case Study Area.	5-37
Figure 5.2-17.	Example of fitted response curve for target ASSETS EI=2 for the Neuse River Estuary.	5-38
Figure 5.2-18.	Theoretical SPARROW response curves demonstrating relative influence of sources on nitrogen loads to an estuary.	5-40
Figure 5.3-1.	Importance of lichens as an indicator of ecosystem health (Jovan, 2008).	5-46
Figure 5.3-2.	Benchmarks of atmospheric nitrogen deposition for several ecosystem indicators.	5-47
Figure 5.3-3.	Areas of highest potential nutrient enrichment sensitivity. (Acidophytic lichens, tree species, and the extent of the Mojave Desert come from data obtained from the United States Forest Service. The extents of coastal sage scrub and California mixed conifer forest come from the California Fire and Resource Assessment Program. Grasslands were obtained from the National Land Cover Dataset [USGS]).	5-57
Figure 5.3-4.	Coastal sage scrub range and total nitrogen deposition using CMAQ 2002 modeling results and NADP monitoring data.	5-61
Figure 5.3-5.	Current fire threats to coastal sage scrub communities.	5-63
Figure 5.3-6.	Mixed conifer forest range and total nitrogen deposition using CMAQ 2002 modeling results and NADP monitoring data.	5-65
Figure 5.3-7.	Presence of acidophyte lichens and total nitrogen deposition in the California mountain ranges using CMAQ 2002 modeling results and NADP monitoring data.	5-70
Figure 5.3-8.	CMAQ 2002 modeling results and NADP monitoring data for deposition of total nitrogen in the western United States.	5-72
Figure 5.3-9.	Observed effects from ambient and experimental atmospheric nitrogen deposition loads in relation to using CMAQ 2002 modeling results and NADP monitoring data. Citations for effect results are from the ISA, Table 4.4 (U.S. EPA, 2008).	5-74

Figure 5.3-10.	Illustration of the range of terrestrial ecosystem effects observed relative to atmospheric nitrogen deposition.	5-75
Figure 5.3-11.	Habitats that may experience ecological benchmarks similar to coastal sage scrub and mixed conifer forest.	5-76
Figure 5.3-12.	Rocky Mountain National Park location relative to the Niwot Ridge Long-Term Ecological Research site and Denver metropolitan area.	5-78
Figure 6.2-1.	The mercury cycle in an ecosystem (USGS, 2006).	6-4
Figure 6.2-2.	Biogeochemical process of mercury methylation.	6-6
Figure 6.2-3.	Distribution pattern in 2006 for state fish consumption advisory listings (U.S. EPA, 2007a).	6-8
Figure 6.2-4.	Spatial and biogeochemical factors influencing methylmercury production.	6-9
Figure 6.2-5.	Preliminary USGS map of mercury methylation-sensitive watersheds derived from more than 55,000 water quality sites and 2,500 watersheds (Myers et al., 2007).	6-10
Figure 6.3-1.	Percentage of total U.S. emissions of greenhouse gases in CO ₂ equivalents (U.S. EPA, 2007b).	6-12
Figure 7-1.	Representation of the benefits assessment process indicating where some ecological benefits may remain unrecognized, unquantified, or unmonetized. (Source: U.S. EPA, 2006).	7-2
Figure 7-2.	Conceptual model showing the relationships among ambient air quality indicators and exposure pathways and the resulting impacts on ecosystems, ecological responses, effects, and benefits to characterize known or anticipated adverse effects to public welfare.	7-4
Figure 7.1-1.	Number of fish species per lake or stream versus ANC level and aquatic status category (colored regions) for lakes in the Adirondack Case Study Area (Sullivan et al., 2006). The five aquatic status categories are described in Table 7.1-1.	7-7
Figure 7.1-2.	ASSETS EI response curve. Point “a” represents the background nitrogen concentration that would occur in the system with no anthropogenic inputs (assuming the system is not naturally eutrophic) or the system at a pristine state. The upper bound of the instream total nitrogen concentration, Point “b,” is the maximum nitrogen concentration at which the system is nitrogen-limited; above this point, the nitrogen inputs to the system no longer affect the eutrophication condition.	7-13
Figure 7.1-3.	Total Atmospheric nitrogen deposition loads for several ecological effects, including California Coastal Sage Scrub, Pacific coast Mixed Conifer Forest, and Pacific Northwest lichen.	7-16

LIST OF TABLES

Table 2.1-1.	Summary of Sensitive Characteristics, Indicators, Effects, and Impacted Ecosystem Services Analyzed for Each Case Study Evaluated in This Review.	2-3
--------------	--	-----

Table 2.4-1.	Ecological Impacts Associated with Acidification, Nutrient Enrichment, and Increased Mercury Methylation and Their Associated Ecosystem Services.....	2-17
Table 3.3-1.	Annual Total Reactive Nitrogen Deposition (kg N/ha/yr) and Sulfur Deposition (kg S/ha/yr) in 2002 for Each Case Study Area, as Well as the Rocky Mountain National Park.....	3-28
Table 4.2-1.	Aquatic Status Categories.....	4-16
Table 4.2-2.	Estimated Average Concentrations of Surface Water Chemistry at 44 Lakes in the Adirondack Case Study Area Modeled Using MAGIC for Preacidification (1860) and Current (2006) Conditions.....	4-17
Table 4.2-3.	Critical Load Exceedances (Nitrogen + Sulfur Deposition > Critical Load) for 169 Modeled Lakes Within the TIME/LTM and EMAP Survey Programs. “No. Lakes” Indicates the Number of Lakes at the Given Acid Neutralizing Capacity Limit; “% Lakes” Indicates the Total Percentage of Lakes at the Given Acid Neutralizing Capacity Limit.....	4-24
Table 4.2-4.	Model Simulated Average Concentrations for Stream Chemistry at 60 Modeled Streams in the Shenandoah Case Study Area for Preacidification and Current Conditions	4-25
Table 4.2-5.	Critical Load Exceedances (Nitrogen + Sulfur Deposition > Critical Load) for 60 Modeled Streams Within the VTSSS-LTM Monitoring Program in the Shenandoah Case Study Area. “No. Streams” Indicates the Number of Streams at the Given Acid Neutralizing Capacity Limit; “% Streams” Indicates the Total Percentage of Streams at the Given Acid Neutralizing Capacity Limit.....	4-31
Table 4.2-6.	Critical Load Exceedances (Nitrogen + Sulfur Deposition > Critical Load) for the Regional Population of 1,849 Lakes in the Adirondack Case Study Area That Are from 0.5 to 2000 ha in Size and at Least 1 m in Depth. Estimates Are Based on the EMAP Lake Probability Survey of 1991 to 1994.....	4-35
Table 4.3-1.	Summary of Linkages Between Acidifying Deposition, Biogeochemical Processes That Affect Ca ²⁺ , Physiological Processes That Are Influenced by Ca ²⁺ , and Effect on Forest Function.....	4-44
Table 4.3-2.	The Three Indicator (Bc/Al) _{crit} Soil Solution Ratios and Corresponding Levels of Protection to Tree Health and Critical Loads.....	4-57
Table 4.3-3.	Number and Location of USFS FIA Permanent Sampling Plots Used in the Analysis of Critical Loads for Full Ranges of Sugar Maple and Red Spruce.....	4-62
Table 4.3-4.	Ranges of Critical Load Values, by Level of Protection (Bc/Al _{crit}) = 0.6, 1.2, and 10.0) and by State, for the Full Distribution Ranges of Sugar Maple and Red Spruce.....	4-63
Table 4.3-5.	Percentages of Plots, by Protection Level (Bc/Al _{crit}) = 0.6, 1.2, and 10.0) and by State, Where 2002 CMAQ/NADP Total Nitrogen and Sulfur Deposition Was Greater Than the Critical Loads for Sugar Maple and Red Spruce.....	4-65
Table 5.2-1.	Value of Commercial Landings for Selected Species in 2007 (Chesapeake Bay Region)	5-11

Table 5.2-2.	Typology Group Categorizations	5-30
Table 5.2-3.	Summary Statistics for Target Eutrophication Index Scenarios — Potomac Estuary	5-35
Table 5.2-4.	Summary Statistics for Target Eutrophication Index Scenarios — Neuse River Estuary	5-38
Table 5.3-1.	Coastal Sage Scrub Ecosystem Area and Total Nitrogen Deposition	5-59
Table 5.3-2.	Mixed Conifer Forest Ecosystem Area and Nitrogen Deposition	5-69
Table 7.1-1.	Aquatic Status Categories	7-8
Table 7.1-2.	Number and Percentage of Lakes in the Adirondack Case Study Area or Streams in the Shenandoah Case Study Area Currently Exceeding Maximum Depositional Loads Required to Maintain a Given ANC	7-9
Table 7.1-3.	Summary of Linkages among Acidifying Deposition, Biogeochemical Processes that Affect Ca ²⁺ , Physiological Processes That Are Influenced by Ca ²⁺ , and Effect on Forest Function	7-11
Table 7.1-4.	Percent of forest plots in the range of sugar maple and res spruce currently exceeding critical loads required to maintain a given Bc/A1.	7-12
Table 7.3-1.	Summary of Information Assessed in the Risk and Exposure Assessment to Aid in Informing Policy Based on Welfare Effects.	7-26

[This page intentionally left blank.]

ACRONYMS AND ABBREVIATIONS

1		
2	Al	aluminum ^{2+,3+}
3	AM	arbuscular mycorrhizae
4	AN	acid anions (NO ₃ ⁻ and SO ₄ ²⁻)
5	ANC	acid neutralizing capacity
6	AQCD	Air Quality Criteria Document
7	ASSETS EI	Assessment of Estuarine Trophic Status eutrophication index
8	Bc	base cation (Ca ²⁺ + K ⁺ + Mg ²⁺)
9	BC	base cation (Ca ²⁺ + K ⁺ + Mg ²⁺ + Na ⁺)
10	Bc _u	base cation (Ca ²⁺ + K ⁺ + Mg ²⁺) uptake
11	Bc _w	base cation (Ca ²⁺ + K ⁺ + Mg ²⁺) weathering
12	BC _w	base cation (Ca ²⁺ + K ⁺ + Mg ²⁺ + Na ⁺) weathering
13	Bc/Al	base cation to aluminum ratio
14	(Bc/Al) _{crit}	base cation to aluminum ratio (indicator)
15	C	carbon
16	Ca ²⁺	calcium
17	CAA	Clean Air Act
18	CAAA	Clean Air Act Amendments
19	CAF	Coastal Assessment Framework
20	CAFO	confined animal feeding operation
21	CAIR	Clean Air Interstate Rule
22	CAL FIRE	California Department of Forestry and Fire Protection
23	CASAC	Clean Air Scientific Advisory Committee
24	CASTNET	Clean Air Status and Trends Network
25	CH ₄	methane
26	Cl ⁻	chloride
27	CLF	critical load function
28	cm	centimeter
29	CMAQ	Community Multiscale Air Quality
30	CO ₂	carbon dioxide
31	CSS	coastal sage scrub
32	DFO	Determined Future Outlook
33	DIN	dissolved inorganic nitrogen
34	DL	depositional load
35	DO	dissolved oxygen
36	DOI	U.S. Department of the Interior
37	EC	ecosystem carbon content
38	EES	Ecological Effects Subcommittee
39	EGU	electric generating unit
40	EMAP	Environmental Monitoring and Assessment Program
41	EPA	U.S. Environmental Protection Agency
42	ESRI	Environmental Systems Research Institute, Inc.
43	FASOMGHG	Forest and Agriculture Sector Optimization Model – Greenhouse Gas
44		version

1	FHWAR	Fishing, Hunting, and Wildlife-Associated Recreation
2	FIA	Forest Inventory and Analysis
3	GHG	greenhouse gas
4	GIS	geographic information systems
5	GPP	gross primary productivity
6	H ⁺	hydrogen ion
7	H ₂ O	water
8	H ₂ SO ₄	sulfuric acid
9	ha	hectare
10	HAB	harmful algal bloom
11	HBEF	Hubbard Brook Experimental Forest
12	HFC	hydrofluorocarbon
13	Hg ²⁺	divalent mercury
14	Hg ⁰	elemental mercury
15	HNO ₃	nitric acid
16	HONO	nitrous acid
17	HUC	hydrologic unit code
18	ICP	International Cooperative Programme
19	IDW	inverse distance weighted
20	IPCC	Intergovernmental Panel on Climate Change
21	IPM	Integrated Planning Model
22	ISA	Integrated Science Assessment
23	K ⁺	potassium
24	KEF	Kane Experimental Forest
25	kg	kilogram
26	kg/ha/yr	kilograms per hectare per year
27	K _{gibb}	gibbsite equilibrium constant
28	km	kilometer
29	LTER	Long-Term Ecological Research
30	LTM	Long-Term Monitoring
31	m	meters
32	MAGIC	Model of Acidification of Groundwaters in Catchments
33	MAHA	Mid-Atlantic Highlands Assessment
34	MCF	mixed conifer forest
35	MCIP	Meteorology-Chemistry Interface Processor
36	MEA	Millennium Ecosystem Assessment
37	mg/L	milligrams per liter
38	Mg ²⁺	magnesium
39	MSA	metropolitan statistical area
40	N	nitrogen
41	N _{de}	denitrification
42	N _i	nitrogen immobilization
43	N _r	total reactive nitrogen
44	N _{ret}	retention of nitrogen
45	N _u	nitrogen uptake
46	N ₂	nitrogen gas

1	N ₂ O	nitrous oxide
2	N ₂ O ₃	nitrogen trioxide
3	N ₂ O ₄	nitrogen tetroxide
4	N ₂ O ₅	dinitrogen pentoxide
5	Na ⁺	sodium
6	NAAQS	National Ambient Air Quality Standards
7	NADP	National Atmospheric Deposition Program
8	NAPAP	National Acid Precipitation Assessment Program
9	NAWQA	National Water Quality Assessment
10	NEE	net ecosystem exchange
11	NEEA	National Estuarine Eutrophication Assessment
12	NEI	National Emissions Inventory
13	NEP	net ecosystem productivity
14	NH ₃	ammonia
15	NH ₄ ⁺	ammonium
16	NH ₄ NO ₃	ammonium nitrate
17	(NH ₄) ₂ SO ₄	ammonium sulfate
18	NH _x	reduced nitrogen
19	NLCD	National Land Cover Data
20	NO	nitric oxide
21	NO ₂	nitrogen dioxide
22	NO ₂ ⁻	nitrite
23	NO ₃ ⁻	nitrate
24	NO _x	nitrogen oxides
25	NO _y	total oxidized nitrogen
26	NOAA	National Oceanic and Atmospheric Administration
27	NPP	net primary productivity
28	NRC	National Research Council
29	NSRE	National Survey on Recreation and the Environment
30	NSWS	National Surface Water Survey
31	NTN	National Trends Network
32	NTR	organic nitrate
33	O ₂	oxygen
34	O ₃	ozone
35	OAQPS	Office of Air Quality Planning and Standards
36	OEC	Overall Eutrophic Condition
37	OH ⁻	hydroxide
38	OHI	Influencing Factors/Overall Human Influence
39	ORD	Office of Research and Development
40	PAN	peroxyacetyl nitrates
41	PFC	perfluorocarbons
42	PM	particulate matter
43	PM _{2.5}	fine particulate matter less than 2.5 microns in size
44	ppb	parts per billion
45	ppm	parts per million
46	ppt	parts per trillion

1	REMAP	Regional Environmental Monitoring and Assessment Program
2	RFNRP	Regional Forest Nutrition Research Project
3	RSM	response-surface model
4	S	sulfur
5	S _{ret}	retention of sulfur
6	S ₂ O	disulfur monoxide
7	S ₂ O ₃ ²⁻	thiosulfate
8	S ₂ O ₇ ²⁻	sulfur heptoxide
9	SAB	Science Advisory Board
10	SAV	submerged aquatic vegetation
11	SF ₆	sulfur hexafluoride
12	Si	silicon
13	SMB	Simple Mass Balance
14	SO	sulfur monoxide
15	SO ₂	sulfur dioxide
16	SO ₃	sulfur trioxide
17	SO ₄ ²⁻	sulfate
18	SO _x	sulfur oxides
19	SOM	soil organic matter
20	SPARROW	SPATIally Referenced Regressions on Watershed Attributes
21	SRB	sulfate-reducing bacteria
22	SSURGO	Soil Survey Geographic Database
23	STORET	STORage and RETrieval
24	TIME	Temporally Integrated Monitoring of Ecosystems
25	TN	total nitrogen
26	TN _{atm}	total nitrogen atmospheric loading
27	TN _s	instream total nitrogen concentration
28	TP	total phosphorus
29	U.S. EPA	U.S. Environmental Protections Agency
30	USFS	United States Forest Service
31	USGS	U.S. Geological Survey
32	VIF	variance inflation factor
33	VOC	volatile organic carbon
34	WTP	willingness to pay
35	µeq/L	microequivalent per liter
36	µg/g	microgram per gram
37	µg/m ₃	microgram per cubic meter
38	µM	micromolar
39		
40		
41		
42		

KEY TERMS

- 1
- 2 **Acid Neutralizing Capacity:** A key indicator of the ability of water to neutralize the acid or
3 acidifying inputs it receives. This ability depends largely on associated biogeophysical
4 characteristics, such as underlying geology, base cation concentrations, and weathering
5 rates.
- 6 **Acidification:** The process of increasing the acidity of a system (e.g., lake, stream, forest soil).
7 Atmospheric deposition of acidic or acidifying compounds can acidify lakes, streams,
8 and forest soils.
- 9 **Adverse Effect:** The response or component of an ecosystem that is deemed harmful in its
10 function.
- 11 **Air Quality Indicator:** The substance or set of substances (e.g., fine particulate matter [PM_{2.5}],
12 nitrogen dioxide [NO₂], sulfur dioxide [SO₂]) occurring in the ambient air for which the
13 National Ambient Air Quality Standards (NAAQS) set a standard level and monitoring
14 occurs.
- 15 **Alpine:** The biogeographic zone made up of slopes above the tree line, characterized by the
16 presence of rosette-forming herbaceous plants and low, shrubby, slow-growing woody
17 plants.
- 18 **Arid Region:** A land region of low rainfall, where “low” is widely accepted to be less than
19 250 millimeters (mm) of precipitation per year.
- 20 **Assessment Endpoint:** An ecological entity and its attributes that are considered welfare effects,
21 as defined in Clean Air Act Section 302(h), and that are analyzed in the assessment.
- 22 **ASSETS rating High:** Low pressure from influencing factors, low overall eutrophic condition
23 OEC, and any expected improvement or no future change in eutrophic condition.
- 24 **ASSETS rating Good:** Low to moderate pressure, low to moderate-low eutrophic condition, and
25 any expected future change in condition.

1 **ASSETS rating Moderate:** Any pressure, moderate-low to moderate-high eutrophic condition,
2 and any expected future change in eutrophic condition.

3 **ASSETS rating Poor:** Moderate-low to high pressure, moderate to moderate-high eutrophic
4 condition, and any expected future change in condition.

5 **ASSETS rating Bad:** Moderate to high pressure, moderate-high to high eutrophic condition, and
6 any expected future change in eutrophic condition.

7 **ASSETS rating Unknown:** Insufficient data for analysis.

8 **Atmospheric Deposition Transformation Function:** Process by which ambient atmospheric
9 concentrations of NO_x and SO_x are translated into a nitrogen and sulfur deposition metric.

10 **Base Cation Saturation:** The degree to which soil cation exchange sites are occupied with base
11 cations (e.g., Ca^{2+} , Mg^{2+} , K^+) as opposed to Al^{3+} and H^+ . Base cation saturation is a
12 measure of soil acidification, with lower values being more acidic. There is a threshold
13 whereby soils with base saturations less than 20% (especially between 10% to 20%) are
14 extremely sensitive to change.

15 **Biologically Relevant Indicator:** A physical, chemical, or biological entity/feature that
16 demonstrates a consistent degree of response to a given level of stressor exposure and
17 that is easily measured/quantified to make it a useful predictor of biological,
18 environmental, or ecological risk.

19 **Critical Load:** A quantitative estimate of an exposure to one or more pollutants, below which
20 significant harmful effects on specified sensitive elements of the environment do not
21 occur, according to present knowledge.

22 **Denitrification:** The anaerobic reduction of nitrogen oxides (NO_x ; e.g., nitrate or nitrite) to
23 gaseous nitrogen (e.g., nitrous oxide [N_2O] or gaseous nitrogen [N_2]) by denitrifying
24 bacteria.

25 **Determined Future Outlook:** An ASSETS index meaning a qualitative measure of expected
26 changes in the system.

- 1 **Dry Deposition:** The removal of gases and particles from the atmosphere to surfaces in the
2 absence of precipitation (e.g., rain, snow) or occult deposition (e.g., fog).
- 3 **Ecological Dose:** The concentration of a toxicant that inhibits a microbe-mediated ecological
4 process by a designated percentage; for example, ED50 inhibits 50%.
- 5 **Ecological Effect Function:** Process by which deposition of nitrogen and sulfur is related to a
6 given ecological indicator.
- 7 **Ecological Exposure:** The exposure of a nonhuman organism to an environmental stressor.
- 8 **Ecological Risk:** The likelihood that adverse ecological effects may occur or are occurring as a
9 result of exposure to one or more stressors (U.S. EPA, 1992).
- 10 **Ecological Risk Assessment:** A process that evaluates the likelihood that adverse ecological
11 effects may occur or are occurring as a result of exposure to one or more stressors (U.S.
12 EPA, 1992).
- 13 **Ecosystem:** The interactive system formed from all living organisms and their abiotic (i.e.,
14 physical and chemical) environment within a given area. Ecosystems cover a hierarchy of
15 spatial scales and can comprise the entire globe, biomes at the continental scale, or small,
16 well-circumscribed systems such as a small pond.
- 17 **Ecosystem Benefit:** The value, expressed qualitatively, quantitatively, and/or in economic terms,
18 where possible, associated with changes in ecosystem services that result either directly
19 or indirectly in improved public welfare. Examples of ecosystem benefits that derive
20 from improved air quality include improvements in habitats for sport fish species, the
21 quality of drinking water and recreational areas, and visibility.
- 22 **Ecosystem Function:** The processes and interactions that operate within an ecosystem. Such
23 processes include but are not limited to nutrient flow, energy flow, water dynamics, and
24 the flux of trace gases.
- 25 **Ecosystem Services:** The ecological processes or functions having monetary or nonmonetary
26 value to individuals or society at large. These are (1) supporting services, such as

1 productivity or biodiversity maintenance; (2) provisioning services, such as food, fiber, or
2 fish; (3) regulating services, such as climate regulation or carbon sequestration; and (4)
3 cultural services, such as tourism or spiritual and aesthetic appreciation.

4 **Ecosystem Structure:** Refers to the species composition, distribution, and interactions with
5 some abiotic attributes of the environment s they vary through space and time.

6 **Elasticity:** The percentage of change in the response variable for a 1% change in the input
7 physical or meteorological characteristic.

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

10 **Greenhouse Gas:** Those gaseous constituents of the atmosphere, both natural and
11 anthropogenic, that absorb and emit radiation at specific wavelengths within the spectrum
12 of infrared radiation emitted by the earth's surface, the atmosphere, and clouds. This
13 property causes the greenhouse effect. Water vapor (H₂O), carbon dioxide (CO₂), N₂O,
14 methane (CH₄), and ozone (O₃) are the primary greenhouse gases in the earth's
15 atmosphere. In addition to CO₂, N₂O, and CH₄, the Kyoto Protocol deals with the
16 greenhouse gases sulfur hexafluoride (SF₆), hydrofluorocarbons, and perfluorocarbons.

17 **Key Elements of Secondary National Ambient Air Quality Standards:**

18 **(a) Indicators**

19 **(1) Air Quality Indicator (for secondary NAAQS):** The air pollutant(s) whose
20 concentration(s) in the ambient air is (are) measured for purposes of determining
21 compliance with the NAAQS. This indicator may either be the actual criteria air pollutant
22 listed in the Clean Air Act or an appropriate surrogate. For example, NO₂ is the current
23 indicator for the primary and secondary NO_x NAAQS and represents all NO_x, while the
24 current indicator for the primary and secondary sulfur oxides (SO_x) NAAQS is SO₂,
25 representing all SO_x.

26 **(2) Ecological Indicator:** A characteristic of an ecosystem that can provide quantitative
27 information on its ecological condition. An indicator can be or contribute to a measure of

1 integrity and sustainability. For example, one indicator of increasing acidification effects
2 in an aquatic ecosystem is a decrease in acid neutralizing capacity (ANC). As a result, a
3 reduction in ANC can lead to acidification of stream water, and thereby, to changes to
4 fish community structure, a good indicator of overall stream health.

5 **(b) Level (of secondary NAAQS):** The specified value of the indicator or metric (see
6 definition below) that is judged requisite to protect the public welfare from any known or
7 anticipated adverse effects associated with the presence of the criteria pollutant in
8 ambient air. The current level of the secondary NO₂ NAAQS indicator is 0.053 parts per
9 million (ppm) (same as primary). The current level of the secondary SO₂ NAAQS
10 indicator is 0.5 ppm. The level of the W126 metric proposed in the 2007 O₃ secondary
11 NAAQS proposal was 21 ppm-hrs.

12 **(c) Averaging Time (for secondary NAAQS):** The period of time over which exposure
13 to metric values at or above the level of the standard is considered relevant. Over that
14 time period, concentrations are averaged or cumulated to determine whether the level of
15 the standard has been met. Examples include 3-hour, 8-hour, 24-hour, seasonal, or annual
16 averages. The current averaging time for the secondary NO₂ NAAQS is a year. The
17 current averaging time for the secondary SO₂ NAAQS is 3 hours.

18 **(d) Form (of secondary NAAQS):** The statistical characteristics of a standard that
19 determine the stringency, stability, and robustness of that standard when implemented.
20 For example, the current secondary O₃ standard is set at the level of 0.075 ppm, averaged
21 over an 8-hour period. To attain this standard, however, only the 3-year average of the
22 fourth-highest daily maximum (rather than the maximum itself) 8-hour average O₃
23 concentrations measured at each monitor within an area over each year is compared to the
24 level of the standard and must not exceed 0.075 ppm. The current form of the secondary
25 NO₂ NAAQS is the annual arithmetic mean. The current form of the secondary SO₂
26 NAAQS is not to be exceeded more than once per year.

27 **Maximum Depositional Load:** The maximum amount of nitrogen and/or sulfur deposition that
28 a given ecosystem can receive without the degradation of the ecological indicator for a
29 targeted effect.

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

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

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

9 **Overall Eutrophic Condition:** An ASSETS index meaning an estimate of current eutrophic
10 conditions derived from data for five symptoms known to be linked to eutrophication.

11 **Overall Human Influence:** An ASSETS index meaning physical, hydrologic, and
12 anthropogenic factors that characterize the susceptibility of the estuary to the influences
13 of nutrient inputs (also quantified as part of the index) and eutrophication.

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

17 **Sensitivity:** The degree to which a system is affected, either adversely or beneficially, by an
18 effect of NO_x and/or SO_x pollution (e.g., acidification, nutrient enrichment). The effect
19 may be direct (e.g., a change in growth in response to a change in the mean, range, or
20 variability of nitrogen deposition) or indirect (e.g., changes in growth due to the direct
21 effect of nitrogen consequently altering competitive dynamics between species and
22 decreased biodiversity).

23 **Target Load:** A policy-based metric that takes into consideration such factors as economic costs
24 and time frame for emissions reduction. The target load can be lower than the critical
25 load if a very sensitive area is to be protected in the short term, especially if deposition
26 rates exceed critical loads.

1 **Total Reactive Nitrogen:** All biologically, chemically, and radiatively active nitrogen
2 compounds in the atmosphere and biosphere, such as ammonia gas (NH₃), ammonium
3 ion (NH₄⁺), nitric oxide (NO), reduced nitrite (NO₂), nitric acid (HNO₃), N₂O, reduced
4 nitrate (NO₃⁻), and organic compounds (e.g., urea, amines, nucleic acids).

5 **Uncertainty:** A measure of the knowledge of the magnitude of a parameter. Uncertainty can be
6 reduced by research (i.e., the parameter value can be refined). Uncertainty is quantified as
7 a distribution. For example, the volume of a lake may be estimated from its surface area
8 and an average depth. This estimate can be refined by measurement (Webster and
9 MacKay, 2003).

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

13 **Variability:** The degree to which values in a distribution differ from each other. Variability can
14 be measured as range, mean, variance and standard deviation.

15 **Variable Factors:** Influences that, by themselves or in combination with other factors, may alter
16 the effects of an air pollutant on public welfare [Clean Air Act Section 108 (a)(2)].

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

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

25 **Vulnerability:** The degree to which a system is susceptible to and unable to cope with the
26 adverse effects of NO_x and/or SO_x air pollution.

1 **Welfare Effects:** The effects on soils, water, crops, vegetation, manmade materials, animals,
2 wildlife, weather, visibility, and climate, damage to and deterioration of property, and
3 hazards to transportation, as well as effects on economic values and on personal comfort
4 and well-being, whether caused by transformation, conversion, or combination with other
5 air pollutants. [Clean Air Act Section 302(h)].

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

8

9

1 **RISK AND EXPOSURE ASSESSMENT FOR REVIEW**
2 **OF THE SECONDARY NATIONAL AMBIENT AIR QUALITY**
3 **STANDARDS FOR OXIDES OF NITROGEN AND**
4 **OXIDES OF SULFUR**

5 **EXECUTIVE SUMMARY**

6 **INTRODUCTION**

7 The U.S. Environmental Protection Agency (EPA) is conducting a joint review of the
8 existing secondary (welfare-based) National Ambient Air Quality Standards (NAAQS) for
9 nitrogen oxides (NO_x) and sulfur oxides (SO_x).¹ EPA has decided to jointly assess the scientific
10 information, associated risks, and standards relevant to protecting the public welfare from
11 adverse effects associated with NO_x and SO_x because NO_x, SO_x, and their associated
12 transformation products are linked from an atmospheric chemistry perspective, as well as an
13 environmental effects perspective, and because the National Research Council (NRC) has
14 recommended that EPA consider multiple pollutants, as appropriate. This is the first time since
15 the NAAQS were established in 1971 that a joint review of these two pollutants has been
16 conducted.

17 **OVERVIEW OF NITROGEN AND SULFUR IN THE**
18 **ENVIRONMENT**

19 Under Section 108 of the Clean Air Act,
20 the secondary standard is to specify an
21 acceptable level of the criteria pollutant(s) in
22 the ambient air that is protective of known or
23 anticipated adverse effects to public welfare.
24 For this review, the relevant atmospheric
25 indicators are ambient NO_x and SO_x
26 concentrations that can be linked to levels of

The sum of mono-nitrogen oxides, nitrogen dioxide (NO₂) and nitric oxide (NO), typically are referred to as nitrogen oxides (NO_x) in the atmospheric science community. More formally, the family of NO_x includes any gaseous combination of nitrogen and oxygen (e.g., NO₂, NO, nitrous oxide [N₂O], nitrogen trioxide [N₂O₃], nitrogen tetroxide [N₂O₄], and dinitrogen pentoxide [N₂O₅]).

Sulfur dioxide (SO₂) is one of a group of substances known as oxides of sulfur, or SO_x, which include multiple gaseous substances (e.g., SO₂, sulfur monoxide [SO], sulfur trioxide [SO₃], thiosulfate [S₂O₃], and heptoxide [S₂O₇], as well as particulate species, such as ammonium sulfate [(NH₄)₂SO₄]).

¹ EPA is also conducting independent reviews of the primary (health-based) NAAQS for NO_x and SO_x. For documents related to this review, see <http://www.epa.gov/ttn/naaqs/standards/no2so2sec/index.html>.

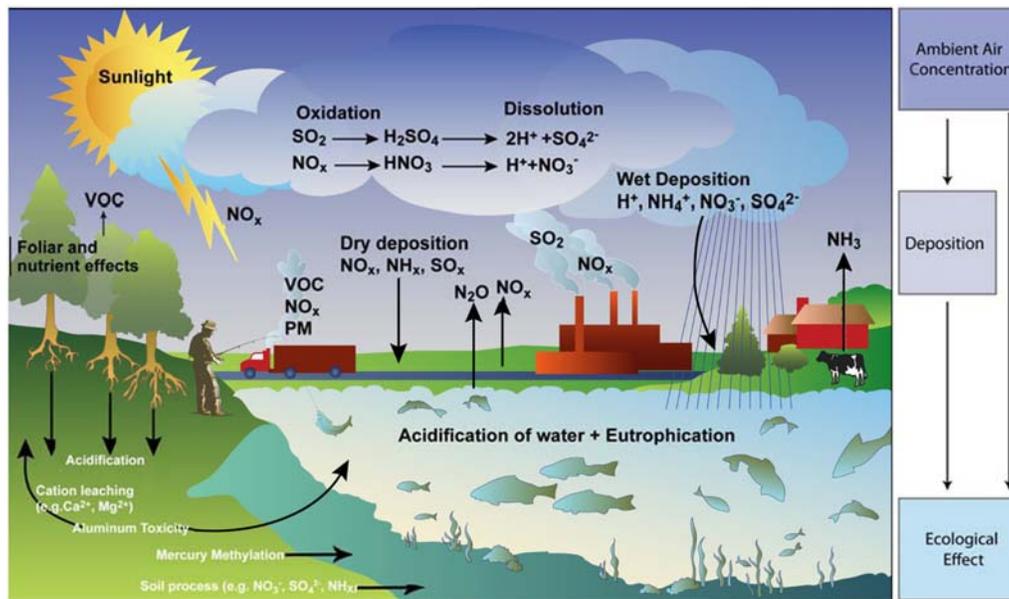
1 deposition for which there are known or anticipated adverse ecological effects. The ecological
2 effects of nitrogen and sulfur are caused both by the gas-phase and atmospheric deposition of the
3 pollutants. The current secondary NAAQS were set to protect against direct damage to
4 vegetation by exposure to gas-phase NO_x or SO_x, such as foliar injury, decreased photosynthesis,
5 and decreased growth.

6 Deposition of nitrogen- and sulfur-containing compounds that are derived from NO_x and
7 SO_x may be wet (e.g., rain, snow), occult (e.g., cloud and fog), or dry (e.g., gases and particles)
8 and can affect ecosystem biogeochemistry, structure, and function. Nitrogen and sulfur
9 interactions in the environment are highly complex. Both are essential, and sometimes limiting,
10 nutrients needed for growth and productivity. Excess nitrogen (both oxidized and reduced forms)
11 or sulfur can lead to acidification, nutrient enrichment, and eutrophication. Acidification causes a
12 cascade of effects that alter both terrestrial and aquatic ecosystems. These effects include slower
13 biomass growth, the injury or death of forest vegetation, and localized extinction of fish and
14 other aquatic species. In addition to acidification, NO_x acts with other forms of reactive nitrogen
15 (including reduced nitrogen) to increase the total amount of available nitrogen in ecosystems.

16 Nitrogen deposition alone alters numerous biogeochemical indicators, including primary
17 productivity that leads to changes in community composition and eutrophication. In aquatic
18 ecosystems, alterations in freshwater lake diatom communities and impaired water quality in the
19 western United States have been observed. In estuarine ecosystems, additional nitrogen from
20 anthropogenic atmospheric sources contributes to the total nitrogen loading and to increased
21 phytoplankton and algal productivity, which leads to eutrophication.

22 In terrestrial ecosystems, nitrate leaching is a well-documented effect that indicates the
23 ecosystem is receiving more nitrogen than it uses. Nitrogen deposition also affects primary
24 productivity, thereby altering terrestrial carbon cycling. This may result in shifts in population
25 dynamics, species composition, community structure, and in extreme instances, ecosystem type.
26 Lichen are the most nitrogen sensitive terrestrial taxa, with documented adverse effects in the
27 Pacific Northwest and in Southern California. Declining biodiversity within grasslands due to
28 nitrogen deposition has also been observed in the central United States.

1 A summary illustration of NO_x and SO_x effects on the environment is presented in **Figure**
 2 **ES-1.**



3
 4 **Figure ES-1.** Nitrogen and sulfur cycling and interactions in the environment.

5 **POLICY-RELEVANT QUESTIONS**

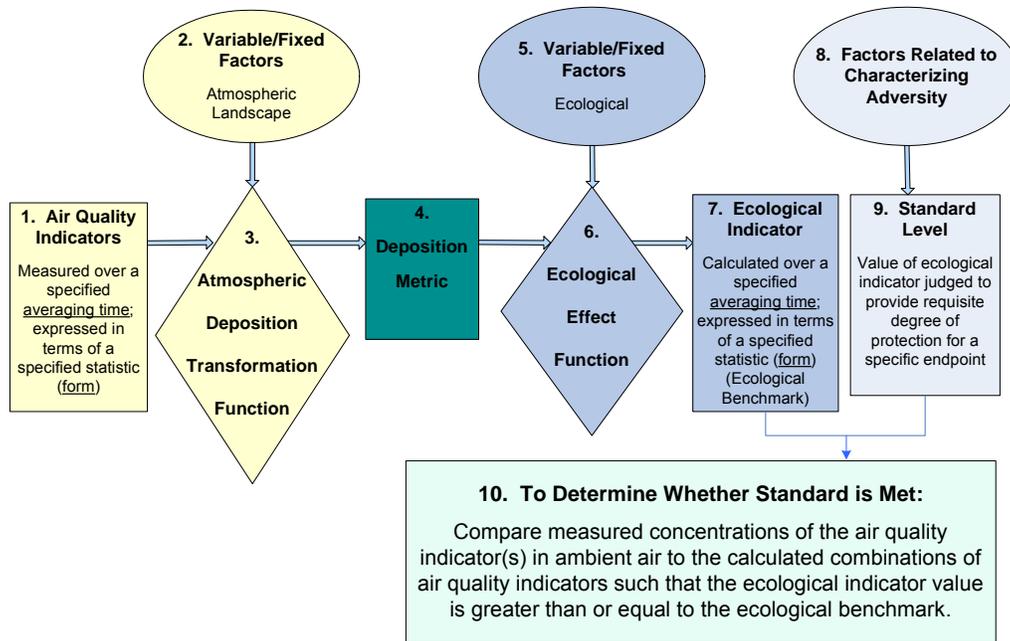
6 To the extent the evidence suggests that the current standards do not provide appropriate
 7 protection from known or anticipated adverse public welfare effects associated with the criteria
 8 pollutants NO_x and SO_x, ecologically meaningful revisions to the current standards will be
 9 considered. Recognizing the high degree of complexity that exists in relationships between
 10 ambient air concentrations of NO_x and SO_x, deposition of nitrogen and sulfur into sensitive
 11 aquatic and terrestrial ecosystems, and associated potential adverse ecological effects, it is
 12 anticipated that ecologically meaningful NAAQS need to be structured to take into account such
 13 complexity. For this secondary NO_x/SO_x NAAQS review, the main policy-relevant questions
 14 include the following:

- 15 ■ To what extent do the current standards provide protection from the known or anticipated
 16 welfare effects associated with NO_x and SO_x?
- 17 ■ To what extent does the current NO_x standard provide protection against known or
 18 anticipated adverse effects associated with total reactive nitrogen?
- 19 ■ What is the nature and magnitude of ecosystem responses to total reactive nitrogen (to
 20 which NO_x contributes) and SO_x that are understood to have known or anticipated
 21 detrimental public welfare effects, and what is the variability associated with those

- 1 responses (including ecosystem type, climatic conditions, and interactions with other
2 environmental factors and pollutants)?
- 3 ■ To what extent can ecological effects due to NO_x be distinguished from effects due to total
4 reactive nitrogen?
 - 5 ■ Does the available information provide support for considering different air quality
6 indicators for NO_x and SO_x?
 - 7 ■ For which ecological effects being considered is a joint NO_x/SO_x standard most
8 appropriate, and for which effects would separate standards be more appropriate?
 - 9 ■ Taking into consideration factors related to determining when the various detrimental
10 ecological effects under consideration occur, what range of levels, averaging times, and
11 forms of alternative ecological indicators are supported by the information, and what are
12 the uncertainties and limitations in that information?
 - 13 ■ To what extent do specific levels, averaging times, and forms of alternative ecological
14 indicators reduce detrimental impacts attributable to NO_x/SO_x relative to current
15 conditions, and what are the uncertainties in the estimated reductions?

16 As many years of research have clearly demonstrated, the ecological effects associated
17 with acidification and nutrient enrichment derive from both oxidized and reduced nitrogen, not
18 “oxides of nitrogen” alone, which is the currently listed criteria pollutant. The policy-relevant
19 questions driving this review recognize that the effects of NO_x occur as part of the overall effects
20 of total reactive nitrogen and address the need to understand the role of NO_x relative to other
21 sources of reactive nitrogen that contribute to adverse public welfare effects. Throughout both
22 the *Integrated Science Assessment (ISA) for Oxides of Nitrogen and Sulfur—Ecological Criteria*
23 *(Final Report)* (ISA) (EPA, 2008) and the Risk and Exposure Assessment, public welfare effects
24 due to total reactive nitrogen are examined, and, where possible, the contributions to these
25 effects from oxidized and reduced forms of nitrogen are assessed.

26 To provide some context for addressing the key policy-relevant questions that are salient
27 in this review, a possible structure has been developed for establishing secondary standards
28 based on meaningful ecological indicators that provides for protection against the range of
29 potentially adverse ecological effects that are associated with the deposition of NO_x, NH_x, and
30 SO_x as shown in **Figure ES-2**. In so doing, consideration has been given as to how the basic
31 elements of NAAQS standards—indicator, averaging time, form, and level—would be reflected
32 in such a structure.



1
2 **Figure ES-2.** Possible structure of a secondary NAAQS for NO_x and SO_x based on an
3 ecological indicator.

4 The framework shown in **Figure ES-2** attempts to depict how an ecologically meaningful
5 secondary standard might be structured. It is a system of linked functions that translates an
6 atmospheric indicator (e.g., concentrations of NO_x and SO_x) into an ecological indicator that
7 expresses either the potential for deposition of nitrogen and sulfur to acidify an ecosystem or for
8 nitrogen to adversely enrich an ecosystem. This encompasses the linkages between ambient air
9 concentrations and resulting deposition metrics, and between the deposition metric and the
10 ecological indicator of concern. The Atmospheric Deposition Transformation Function (box 3)
11 translates ambient atmospheric concentrations of NO_x and SO_x to nitrogen and sulfur deposition
12 metrics, while the Ecological Effect Function (box 6) relates the deposition metric into the
13 ecological indicator.

14 The amounts of NO_x and SO_x in the ambient atmosphere can be used to derive a
15 deposition metric (via the atmospheric deposition transformation function) that can then be used
16 to derive a level of an ecological indicator (through the ecological effect function), which falls
17 within the range defined as acceptable by the standard, and by definition, the levels of NO_x and
18 SO_x will be considered to meet that standard of protection. The maximum amount of nitrogen
19 and/or sulfur deposition that a given ecosystem can receive without the degradation of the
20 ecological indicator for targeted effects is referred to as the maximum deposition load.

RISK AND EXPOSURE ASSESSMENT APPROACH

Because ecosystems are diverse in biota, climate, geochemistry, and hydrology, response to pollutant exposures can vary greatly between ecosystems. This Risk and Exposure Assessment addresses four main ecosystem effects identified in the 2008 ISA:

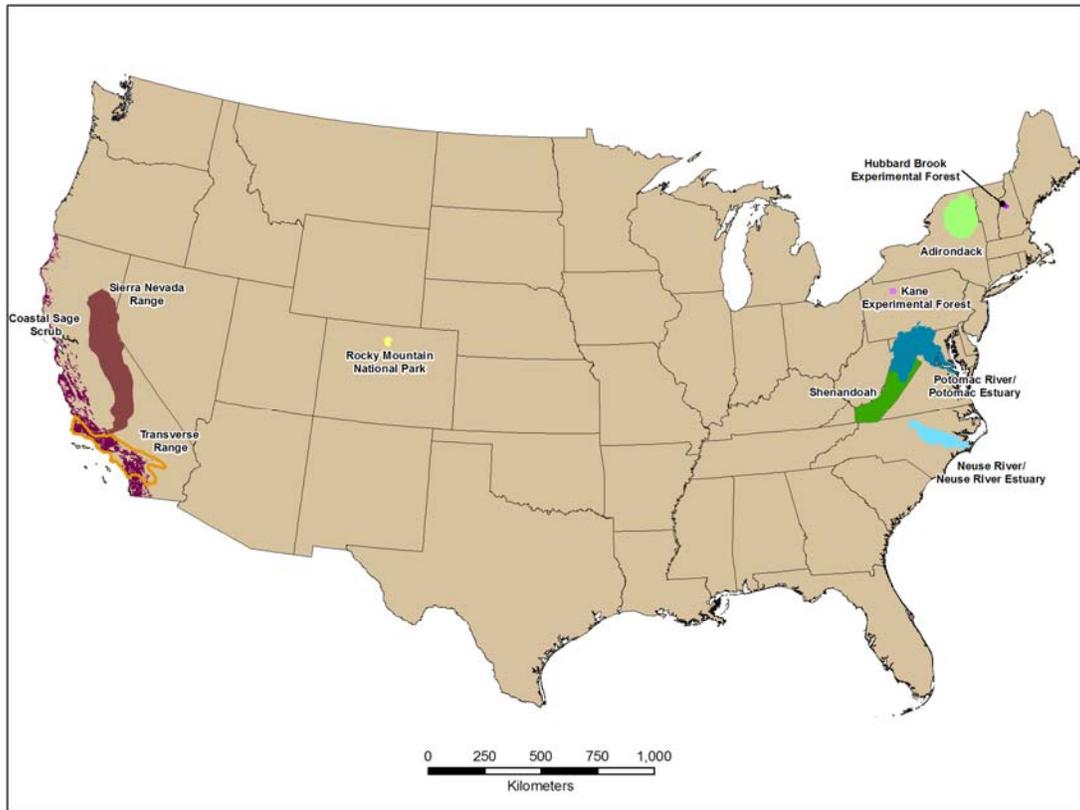
- Aquatic acidification due to nitrogen and sulfur
- Terrestrial acidification due to nitrogen and sulfur
- Aquatic nitrogen enrichment, including eutrophication
- Terrestrial nitrogen enrichment.

Because these ecosystem effects are not evenly distributed across the United States, case studies have been developed for these analyses based on ecosystems identified as sensitive to nitrogen and/or sulfur deposition effects. This assessment builds upon the scientific information presented in the ISA (U.S. EPA, 2008). Ecological indicator(s) and case study locations were selected based on the information presented in the ISA (U.S. EPA, 2008). The case study locations are described in **Table ES-1**, along with a summary of the ecosystem characteristics, indicators, and ecosystem service information regarding these locations that were identified and analyzed for the Risk and Exposure Assessment. A map highlighting each of the case study areas is shown in **Figure ES-3**.

Table ES-1. Summary of Sensitive Characteristics, Indicators, Effects, and Impacted Ecosystem Services Analyzed for Each Case Study Evaluated in This Review

Targeted Ecosystem Effect	Characteristics of Sensitivity (Variable Ecological Factors)	Biological/ Chemical Indicator	Ecological Endpoint	Ecological Effects	Ecosystem Services Impacted	Case Study Areas
Aquatic Acidification	Geology, surface water flow, soil depth, weathering rates	Al pH ANC	Species richness, abundance, composition, ANC	Species losses of fish, phytoplankton, and zooplankton; changed community composition, ecosystem structure, and function	Subsistence fishing, recreational fishing, other recreational activities	Adirondack Mountains, NY (referred to as Adirondack) Shenandoah National Park, VA (referred to as Shenandoah)
Terrestrial Acidification	Geology, surface water flow, soil depth, weathering rates	Soil base saturation Al Ca C:N ratio	Tree health of red spruce and sugar maple, ANC, base cation :Al ratio	Decreased tree growth, increased susceptibility to stress, episodic dieback; changed community composition, ecosystem structure, and function	Provision of food and wood products, recreational activities, natural habitat, soil stabilization, erosion control, water regulation, climate regulation	Kane Experimental Forest (Allegheny Plateau, PA) Hubbard Brook Experimental Forest (White Mountains, NH)
Aquatic Nutrient Enrichment	Nitrogen-limited systems, presence of nitrogen in surface water, eutrophication status, nutrient criteria	Chlorophyll <i>a</i> , macroalgae, dissolved oxygen, nuisance/toxic algal blooms, submerged aquatic vegetation (SAV)	Changes in Eutrophication Index (EI)	Habitat degradation, algal blooms, toxicity, hypoxia, anoxia, fish kills, decreases in biodiversity	Commercial and recreational fishing, other recreational activities, aesthetic value, nonuse value flood and erosion control	Potomac River Basin, Chesapeake Bay (referred to as Potomac River/Potomac Estuary) Neuse River Basin, Pamlico Sound (referred to as Neuse River/Neuse River Estuary)
Terrestrial Nutrient Enrichment	Presence of acidophytic lichens, anthropogenic land cover	Cation exchange capacity, C:N ratios, Ca:Al ratios, NO ₃ ⁻ leaching and export	Species composition, lichen presence/absence, soil root mass changes, NO ₃ breakthrough to water, biomass	Species changes, nutrient enrichment of soil, changes in fire regime, changes in nutrient cycling	Recreation, aesthetic value, nonuse value, fire regulation, loss of habitat, loss of biodiversity, water quality	Coastal Sage Scrub (southern, coastal California) and Mixed Conifer Forest (San Bernardino Mountains of the Transverse Range and Sierra Nevada Mountain Ranges, California); Rocky Mountain National Park (a supplemental study area)

1 **Note:** ANC = acid neutralizing capacity, SAV = submerged aquatic vegetation, EI = eutrophication index.

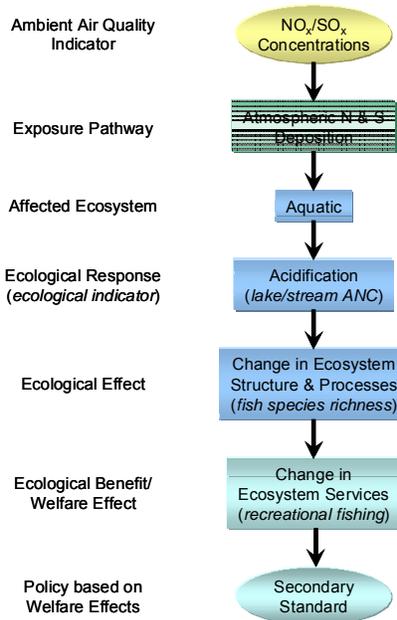


1
2 **Figure ES-3** National map highlighting the 8 case study areas and the Rocky Mountain
3 National Park (a supplemental study area) evaluated in the Risk and Exposure
4 Assessment.

5 For the purposes of assessing this set of secondary NAAQS, in addition to assessing the
6 degree of impairment of ecological systems relating to inputs of NO_x and SO_x , a broad look into
7 the concept of ecosystem services is being made and can help link what is considered to be a
8 biologically adverse effect with a known or anticipated adverse effect to public welfare. In this
9 Risk and Exposure Assessment, ecosystem services is being used as an umbrella term, which can
10 aid in describing the impacts of ecological effects on public welfare. It is a way to help explain
11 how ecosystem effects are viewed by the public. The ability to inform decisions on the level of a
12 secondary NAAQS will require the development of clear linkages between biologically adverse
13 effects and effects that are known or anticipated to be adverse to public welfare through
14 ecosystem services. The concept of adversity to public welfare does not require the use of
15 ecosystem services, yet it is envisioned as a beneficial tool for this review that may provide more
16 information on the linkages between changes in ecological effects and known or anticipated
17 adverse public welfare effects.

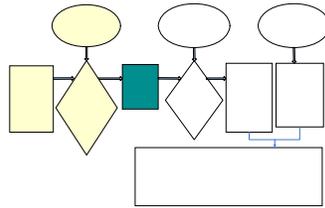
1 As described in the EPA's *Ecological Benefits Assessment Strategic Plan* (U.S. EPA,
 2 2006), it is necessary to recognize that in the analysis of the environmental responses associated
 3 with any particular policy or environmental management action, some of the ecosystem services
 4 likely to be affected are readily identified, while others will remain unidentified. Of those
 5 ecosystem services that are identified, some changes can be quantified, whereas others will
 6 remain unidentified. Within those services whose changes are quantified, only a few will likely
 7 be monetized, and many will remain unmonetized. Similar to health effects, only a portion of the
 8 ecosystem services affected by a policy can be monetized. A conceptual model integrating the
 9 role of ecosystem services in characterizing known or anticipated adverse effects to public
 10 welfare is shown in **Figure ES-4**.

11 Knowledge about the relationships linking ambient concentrations and ecosystem
 12 services can be used to inform a policy judgment on a known or anticipated adverse public
 13 welfare effect. The conceptual model outlined for aquatic acidification in **Figure ES-4** can be
 14 modified for any targeted effect area where sufficient data and models are available. This
 15 information can then be used to characterize known or anticipated adverse effects to public
 16 welfare and to inform a policy based on welfare effects.



17
 18 **Figure ES-4.** Conceptual model showing the relationships among ambient air quality
 19 indicators and exposure pathways and the resulting impacts on ecosystems, ecological
 20 responses, ecological effects, and finally, on the quality of a particular activity (e.g.,
 21 recreational fishing) known to influence public welfare.

NITROGEN AND SULFUR IN THE ATMOSPHERE



The air quality analyses for this review encompass the current emissions sources of nitrogen and sulfur, as well as atmospheric concentrations, estimates of deposition of total nitrogen, policy-relevant background, and nonambient loadings of nitrogen and sulfur to ecosystems, both nationwide and in the case study areas.

EMISSION SOURCES OF NITROGEN AND SULFUR

Annual total emissions for 2002 from the National Emissions Inventory (NEI) were used to characterize the magnitude and spatial patterns in emissions of NO_x , NH_3 , and SO_2 nationwide². NO_x emissions are the largest of these three pollutants, at over 20 MM tons per year, followed by SO_x , at over 16 MM tons per year. Emissions of NH_3 , at over 4 MM tons per year, are relatively small by comparison to emissions of NO_x and SO_x , but they may be important locally.

Anthropogenic sources account for the vast majority of total NO_x emissions (i.e., 60% mobile sources and 40% stationary sources). The primary anthropogenic sources of NH_3 emissions are fertilized soils and livestock, with highest emissions generally found in areas of major livestock feeding and production facilities, most of which are in rural areas.

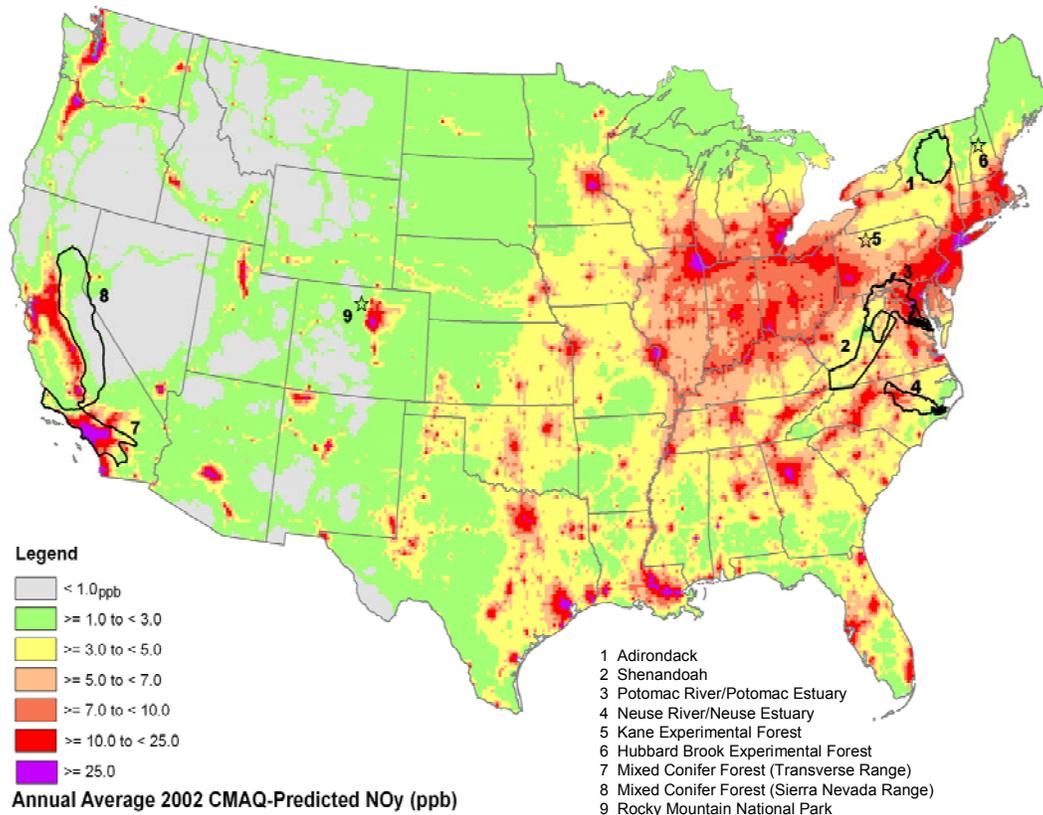
Fossil fuel combustion by electrical utilities emit about 66% of the nation's SO_2 , industrial sources emit about 29%, and mobile sources emit about 5%.

AMBIENT CONCENTRATIONS

Air quality model predictions are taken from applications of the Community Multiscale Air Quality (CMAQ) model. CMAQ was used to simulate concentrations and deposition for 2002 using meteorology and emissions for this year (with a horizontal resolution of approximately 12 x 12 kilometers [km]). **Figures ES-5 and ES-6** show the spatial field of

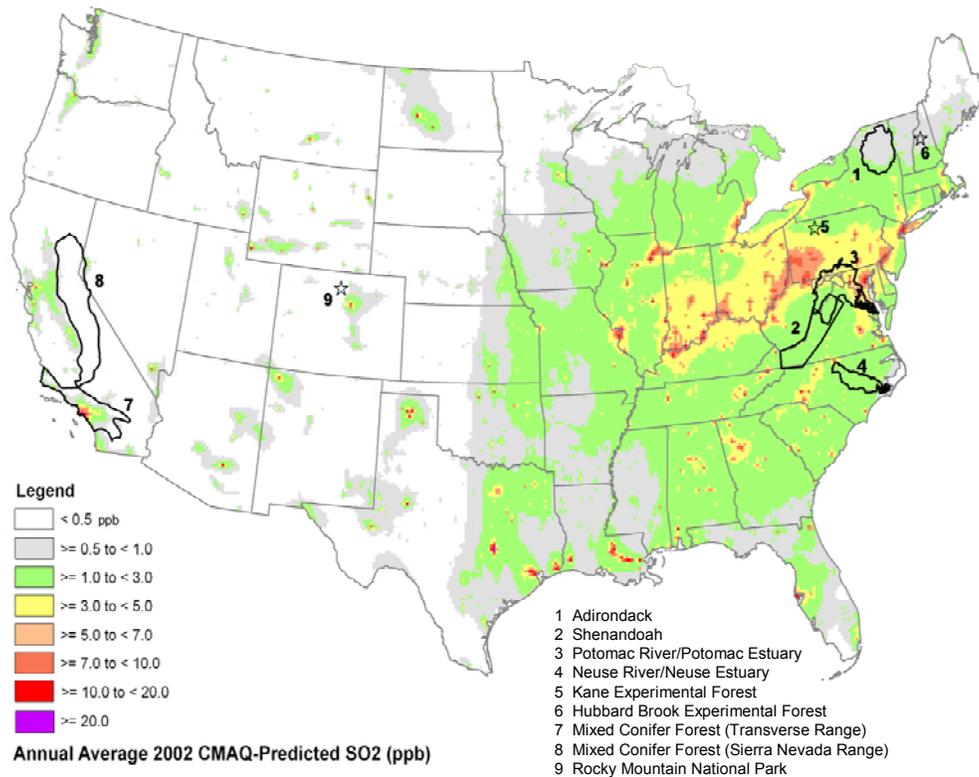
² For the purposes of this analysis, nationwide emissions do not include emissions from Alaska or Hawaii.

1 model-predicted 2002 annual average NO_x and SO₂ concentrations, respectively. The patterns in
 2 NO_x concentrations generally mirror the patterns of NO_x emissions. The model predictions are
 3 generally consistent with the magnitude of concentrations from measured data. Peak SO₂
 4 concentrations, exceeding 10.0 parts per billion (ppb), coincide with the location of highest
 5 emissions, with large decreases in concentrations with distance from sources.



6
 7 **Figure ES-5.** Model-predicted annual average NO_y concentrations (ppb) for 2002.

8

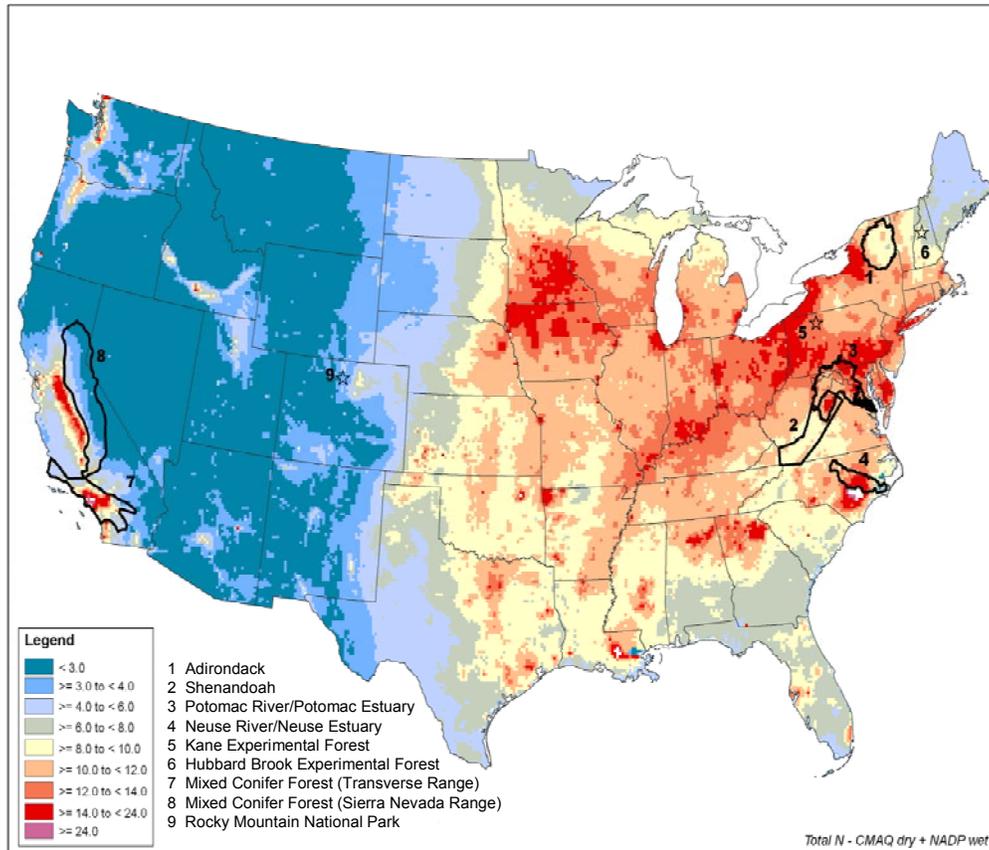


1
2 **Figure ES-6.** Model-predicted 2002 annual average SO₂ concentrations (ppb).

3 **ATMOSPHERIC DEPOSITION**

4 To create spatial fields of deposition, wet deposition measurements from the National
5 Atmospheric Deposition Program (NADP) National Trends Network were used. For dry
6 deposition, predictions from the 2002 CMAQ model simulation were used because the model has
7 information about meteorology and land use in each grid cell of the domain.

8 **Nitrogen Deposition.** The spatial patterns of total nitrogen deposition in **Figure ES-7**
9 reflect the combination of the deposition from the reduced and oxidized nitrogen components.
10 Much of the East has total nitrogen deposition of 9 to 12 kilograms (kg) N/ha/yr. Higher amounts
11 of 12 kg N/ha/yr or greater cover large portions of the Midwest and Northeast and are found in or
12 near sources of NO_x and/or NH₃ emissions in other parts of the East. In the West, total nitrogen
13 deposition is highest in and near NO_x and NH₃ source areas, particularly those in portions of
14 California where deposition exceeds 18 kg N/ha/yr. In most rural/remote portions of the West,
15 total nitrogen deposition is generally less than 3 kg N/ha/yr.



1

2

Figure ES-7. Total reactive nitrogen deposition (kg N/ha/yr) in 2002.

3

4

5

6

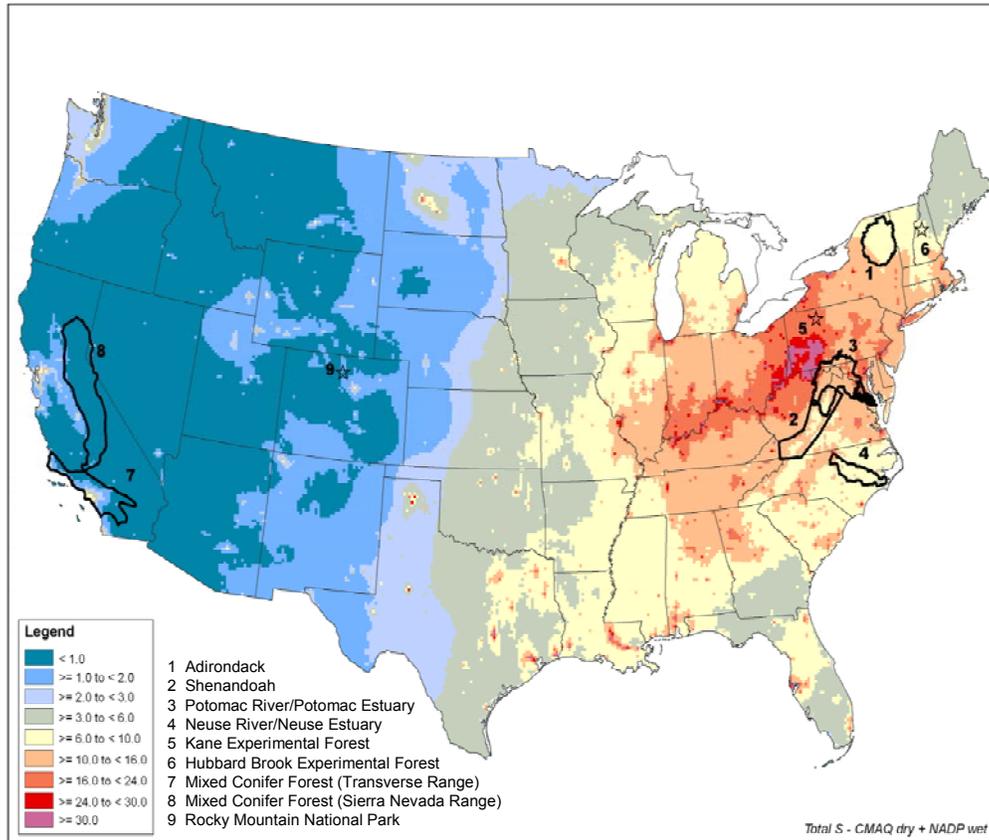
7

8

9

10

Sulfur Deposition. Figure ES-8 shows the spatial fields of sulfur across the United States for 2002. Like deposition of nitrogen species, sulfur deposition is much higher in the East than in the West. Sulfur deposition across most of the West is less than 3.0 kg S/ha/yr. In the East, high levels of deposition exceeding 18 kg S/ha/yr occur in the immediate vicinity of isolated major sources, as well as in and near areas having a high concentration of SO₂ sources. This is particularly notable along the Ohio River Valley extending across Pennsylvania. The areas of highest deposition are within a broad area of sulfur deposition in the range of 6 to 12 kg S/ha/yr, which covers much of the East.

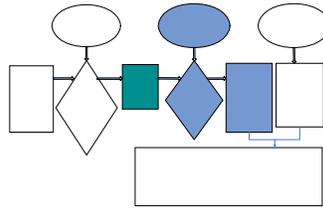


1
2 **Figure ES-8.** Total wet and dry sulfur deposition (kg S/ha/yr) in 2002.

3 **POLICY-RELEVANT BACKGROUND CONCENTRATIONS**

4 Policy-relevant background concentrations are those concentrations that would occur in
5 the United States in the absence of anthropogenic emissions in continental North America
6 (defined here as the United States, Canada, and Mexico). For NO_x, policy-relevant background
7 concentrations are <300 parts per trillion (ppt) over most of the continental United States and
8 <100 ppt in the eastern United States on an annual average basis. Background SO₂
9 concentrations are <10 ppt throughout most of the continental United States, except in areas of
10 the Pacific Northwest, where natural SO₂ sources are particularly strong because of volcanic
11 activity.

ACIDIFICATION



Deposition of SO_x , NO_x , and NH_x leads to ecosystem exposure to acidification. The ISA reported that acidifying deposition has altered major biogeochemical processes in the United States by increasing the sulfur and nitrogen content of soils, accelerating sulfate (SO_4^{2-}) and nitrate (NO_3^-) leaching from soil to drainage water, depleting base cations (especially calcium [Ca^{2+}] and magnesium [Mg^{2+}] from soils, and increasing the mobility of aluminum (Al). Acidification can degrade the health of terrestrial and aquatic ecosystems. One of the effects of soil acidification is the increased mobility of inorganic Al, which is toxic to tree roots, fish, algae, and aquatic invertebrates.

Acidification is the decrease of **acid neutralizing capacity** in water or **base saturation** in soil caused by natural or anthropogenic processes.

Case study analyses on aquatic acidification and terrestrial acidification were performed as part of the Risk and Exposure Assessment to aid in determining whether a link can be established between NO_x and SO_x deposition and ecosystem response. These case studies also tested whether area-based risk and exposure assessments are a suitable method for predicting acidification effects on other ecosystems and geographic regions. The studies facilitate extrapolation of impacts from smaller scale (yet representative) areas to other sensitive areas in the country.

AQUATIC ACIDIFICATION

The changes in ecosystem structure and processes associated with aquatic acidification include changes in fish species richness as measured by the ecological indicator, acid neutralizing capacity (ANC). The impact of acidifying deposition on aquatic systems is controlled by several environmental factors, such as geology, surface water flow, soil depth, and weathering rates, all of which contribute to the ability of a watershed to neutralize the additional acidifying deposition and to prevent the lowering of surface ANC concentrations. ANC is a useful ecological indicator because it integrates the overall acid-base status of a lake or stream and reflects how aquatic ecosystems respond to acidifying deposition over time. There is also a

2 relationship between ANC and the surface water
4 constituents that directly contribute to or
6 ameliorate acidity-related stress, in particular,
8 concentrations of hydrogen ion (as pH), calcium
10 (Ca^{2+}), and Al. In aquatic systems, there is a
12 direct relationship between ANC and fish and

The **critical load approach** provides a means of gauging whether a group of lakes or streams in a given area receives deposition that results in a level of biological harm that is defined by an acid neutralizing capacity (ANC) concentration, known as the critical limit, which corresponds to harmful biological effects (e.g., ANC of 50 $\mu\text{eq/L}$). The greater the critical load value, the greater the ability of the watershed to neutralize the additional acidic anions and protect aquatic life.

13 phyto-zooplankton diversity and abundance (Baker and Brezonik, 1988).MAGIC was used to
14 determine the past (preacidification), present (2002 and 2008), and future (2020 and 2050) acidic
15 conditions of lakes in the Adirondack Case Study Area and streams in the Shenandoah Case
16 Study Area. Furthermore, MAGIC was used to evaluate the associated risk and uncertainty of the
17 current levels of acidification, given the preacidification water quality and the levels of
18 uncertainty in the input parameters. The MAGIC model output for each waterbody was
19 summarized into five ANC levels that correspond to the aquatic status categories: *Acute*
20 *Concern*, *Severe Concern*, *Elevated Concern*, *Moderate Concern*, and *Low Concern* (Sullivan et
21 al., 2006). This grouping offers an assessment of the current risk to the biota of the current
22 condition compared to preacidification and future conditions. Surface water chemistry data were
23 used from two EPA-administered surface water monitoring and survey programs: Temporally
24 Integrated Monitoring of Ecosystems (TIME) and Long-Term Monitoring (LTM). Average
25 yearly ANC concentrations were calculated from annual measurements.

26 The results indicated that approximately 50% of the 169 lakes modeled in the Adirondack
27 Case Study Area are sensitive or at risk to acidifying deposition. For the 2002 model year,
28 maximum depositional loads for ANC values of 0, 20, 50, and 100 $\mu\text{eq/L}$ were calculated. The
29 exceedance value of a maximum depositional load indicates the combined sulfur and nitrogen
30 deposition in year 2002 that is greater than the amount of deposition the lake could buffer and
31 still maintain the ANC level of above each of the four different ANC limits of 0, 20, 50, and 100
32 $\mu\text{eq/L}$. These data were extrapolated for the regional population of 1,849 lakes in the Adirondack
33 Case Study Area that are from 0.5 to 2,000 ha in size and at least 1 meter in depth, based on the
34 Environmental Monitoring and Assessment Program (EMAP) Lake Probability Survey of 1991
35 to 1994. A similar analysis showed that approximately 75% of the 60 streams modeled in the
36 Shenandoah Case Study Area are sensitive or at risk to acidifying deposition. For the year 2002,
37 52%, 72%, 85%, and 92% of the 60 streams modeled received levels of combined sulfur and

1 nitrogen deposition that exceeded maximum depositional loads of 0, 20, 50, and 100 $\mu\text{eq/L}$,
2 respectively. It was not possible to extrapolate the Shenandoah Case Study Area stream data to a
3 larger dataset.

4 The connection between changes in ecological effects associated with declining ANC
5 levels and changes in ecosystem services may aid in determining adverse impacts to public
6 welfare. Examples of these ecosystem services include recreational and subsistence fishing, food,
7 and freshwater.

8 **TERRESTRIAL ACIDIFICATION**

9 Calcium and Al are strongly influenced by soil acidification, and both have been shown
10 to have quantitative links to tree health. The base cation (Bc) to Al ratio (Bc/Al) was used to
11 represent the Ca^{2+}/Al indicator to calculate critical deposition loads of acidity. The Bc variable
12 consists of Ca^{2+} , magnesium (Mg^{2+}), and potassium (K^+), with Ca^{2+} often representing a large
13 proportion of Bc. Sverdrup and Warfvinge (1993) summarized effects based on Bc/Al ratios. At
14 a Bc/Al of 0.6, 75% of tree species showed >20% reduction in fine-root growth, and at a Bc/Al
15 of 1.2, 50% of tree species showed >20% reduction in fine-root growth. These findings
16 demonstrate that as the Bc/Al is reduced, there is a greater likelihood of a negative impact on tree
17 health.

18 The tree species most sensitive to acidification of soils due to atmospheric nitrogen and
19 sulfur deposition include sugar maple (*Acer saccharum*, a deciduous tree species) and red spruce
20 (*Picea rubens*, a coniferous tree species). Much of the scientific literature discussing terrestrial
21 soil acidification focuses on Ca^{2+} depletion and Al mobilization as the primary indicators of
22 detrimental effects to terrestrial vegetation. Both of these indicators are strongly influenced by
23 soil acidification, and both have been shown to have quantitative links to vegetation growth and
24 vigor.

25 Three values of the indicator were used to calculate critical loads of $(\text{Bc}/\text{Al})_{\text{crit}}$, which
26 represent different levels of tree protection associated with total nitrogen and sulfur deposition;
27 0.6, 1.2, and 10. Critical loads for 2002 were calculated for multiple areas within 24 states for
28 sugar maple and in 8 states for red spruce using data from the U.S. Forest Service Forest
29 Inventory and Analysis (FIA) database. The exceedance value of a critical load indicates the
30 combined sulfur and nitrogen deposition in year 2002 that is greater than the amount of

1 deposition forest soils could buffer and still maintain the Bc/Al level of above each of the three
 2 different Bc/Al limits of 0.6, 1.2, and 10. **Table ES-2** summarizes the results.

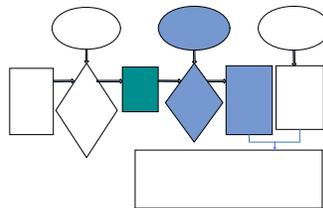
Table ES-2. Percentage of forest plots in the range of sugar maple and red spruce currently exceeding maximum depositional loads required to maintain a given Bc/Al.

	Bc/Al = 0.6	Bc/Al = 1.0	Bc/Al = 10
Sugar Maple (n=4,992; 24 states)	3	12	75
Red Spruce (n=763; 8 states)	3	5	36

3
 4 The influence of negative impacts on tree health by Al/Ca²⁺ imbalances can result in a
 5 decline in ecosystem services. The two main types of products derived from sugar maples are
 6 wood products and maple syrup. The total removal of sugar maple saw timber from timberland
 7 in the United States was almost 900 million board feet in 2006. From 2005 to 2007, annual
 8 production of maple syrup in the United States varied between 1.2 million and 1.4 million
 9 gallons, which accounted for roughly 19% of worldwide production. The total removal of red
 10 spruce saw timber from timberland in the United States was 328 million board feet in 2006. Red
 11 spruce forests are home to the spruce-fir moss spider (endangered), the rock gnome lichen
 12 (endangered), and the Virginia northern flying squirrel (delisted, but considered important).

13 From 1999 to 2004, 16% of adults in the northeastern United States³ participated in off-
 14 road vehicle recreation, with an implied total annual value of more than \$9.25 billion. Further,
 15 the implied total annual values of hunting and wildlife viewing in the northeastern United States
 16 were \$4.38 billion and \$4.21 billion, respectively, in 2006.

17 NUTRIENT ENRICHMENT



18
 19 Nutrient enrichment describes a condition where an increase in a nutrient, such as
 20 nitrogen may result in an imbalance in ecological stoichiometry, causing effects on ecological

³ This area includes Connecticut, Delaware, District of Columbia, Illinois, Indiana, Maine, Maryland, Massachusetts, Michigan, New Hampshire, New Jersey, New York, Ohio, Pennsylvania, Rhode Island, Vermont, West Virginia, and Wisconsin.

1 processes, structure, and function. Some organisms may at first respond positively to an initial
2 increase in nutrients, exhibiting an increase in growth due to fertilization effects. However, as the
3 nutrient load continues to rise, the imbalance can have negative effects either in the organism's
4 response or in the invasion of new organisms that benefit from increased nutrients. In general,
5 ecosystems that are most responsive to nutrient enrichment from atmospheric nitrogen deposition
6 are those that receive high levels of deposition relative to nonanthropogenic nitrogen loading,
7 those that are nitrogen limited, or those that contain species that have evolved in nutrient-poor
8 environments. Nutrient enrichment in ecosystems may alter the native terrestrial species'
9 composition (i.e., species shift from wildflower meadows to shrubs) and can result in
10 eutrophication in aquatic systems.

11 **AQUATIC NUTRIENT ENRICHMENT**

12 Nitrogen is an essential nutrient for aquatic ecosystem fertility, including lake, marine,
13 and estuarine ecosystems, and is often the limiting nutrient for growth and reproduction in many
14 of these ecosystems. Nutrient enrichment may have beneficial fertilization effects but can also
15 lead to over-enrichment of a system, causing eutrophication. Excessive nitrogen enrichment can
16 change ecosystem structure and function by causing harmful algal blooms, hypoxia (i.e., reduced
17 dissolved oxygen), anoxia (i.e., absence of dissolved oxygen), fish kills, habitat degradation, and
18 decreases in biodiversity.

19 There is strong scientific consensus that nitrogen is the principal cause of coastal
20 eutrophication in the United States. On average, human activity has likely contributed to a six-
21 fold increase in the nitrogen flux to U.S. coastal waters, and nitrogen now represents the most
22 significant coastal pollution problem. Atmospheric deposition is responsible for a portion of the
23 nitrogen input. The Aquatic Nutrient Enrichment Case Study for the Risk and Exposure
24 Assessment focuses on two estuarine systems—the Potomac Estuary and the Neuse River
25 Estuary.

26 Due to the cascading impacts and effects of nitrogen enrichment, there are a suite of
27 possible ecological indicators. The National Oceanic and Atmospheric Administration's
28 (NOAA) National Estuarine Eutrophication Assessment (NEEA) Update provides a detailed
29 explanation of the biological indicators used to evaluate eutrophic status. Five biological
30 indicators are used in this index: chlorophyll *a*, macroalgae, dissolved oxygen, nuisance/toxic

1 algal blooms, and submerged aquatic vegetation (SAV). NOAA's Assessment of Estuarine
2 Trophic Status (ASSETS) eutrophication index (EI) is an estimation of the likelihood that the
3 estuary is experiencing eutrophication or will experience eutrophication in the future based on
4 the five indicators listed above.

5 In this assessment, two main stem rivers, the Potomac and the Neuse, were selected to
6 analyze the influence of total atmospheric nitrogen deposition on eutrophic conditions to the
7 Potomac Estuary and Neuse River Estuary, respectively. The ASSETS EI for both of these
8 estuaries is currently "Bad." Response curves were developed that related in-stream nitrogen
9 levels to the ASSETS EI. The curves were used to determine the in-stream nitrogen
10 concentrations necessary to move the ASSETS EI from a score of "Bad" to "Poor," a one-
11 category improvement. The in-stream nitrogen concentrations can be used to back-calculate the
12 required decrease in atmospheric deposition to achieve those concentrations. For both estuaries, a
13 100% or greater reduction in atmospheric deposition was necessary, demonstrating that
14 reductions in additional sources of nitrogen loading to the estuaries are also required.

15 Estuaries are capable of supporting large stocks of resident commercial species, and they
16 serve as the breeding grounds and interim habitat for several migratory species. The ecosystem
17 services impacted by nitrogen loadings to estuaries include commercial and residential fishing,
18 and shoreline protection from erosion and flooding.

19 In addition to the case studies for the Potomac and Neuse River estuaries, the ISA (U.S.
20 EPA, 2008) presents scientific studies that show that increased atmospheric nitrogen deposition
21 in high alpine lakes and streams can cause a shift in community composition and reduce algal
22 biodiversity. Elevated nitrogen deposition results in changes in algal species composition,
23 especially in sensitive oligotrophic lakes. Two opportunistic diatom species, *Asterionella*
24 *formosa* and *Fragilaria crotonensis* (McKnight et al., 1990; Lafrancois et al., 2004; Saros et al.,
25 2005), now dominate the flora of at least several alpine and montane Rocky Mountain lakes,
26 with similar field data showing shifts in dominant algal species in other parts of the West. A
27 hindcasting exercise has concluded that the change in Rocky Mountain National Park lake algae
28 that occurred between 1850 and 1964 was associated with an increase in wet nitrogen deposition
29 that was only about 1.5 kg N/ha (Baron, 2006). Similar changes inferred from lake sediment
30 cores of the Beartooth Mountains of Wyoming also occurred at about 1.5 kg N/ha deposition
31 (Saros et al., 2003). There is a strong relationship between aquatic eutrophication of high alpine

1 lakes in the Rocky Mountains and atmospheric deposition, since atmospheric deposition is the
2 only source of nitrogen to these systems.

3 **TERRESTRIAL NUTRIENT ENRICHMENT**

4 Excess nitrogen in terrestrial ecosystems changes ecosystem structure and processes by
5 inadvertent fertilization of vegetation; creating increased growth rates in some species over
6 others, which changes competitive interactions among species; and nutrient imbalances. These
7 impacts ultimately reduce ecosystem health and biodiversity. Enhanced growth generally occurs
8 above ground level (i.e., stem and leaves), producing more shoot growth compared to root
9 growth. This increase in shoot-to-root ratio can cause decreased resistance to environmental
10 stressors, such as drought. In conifer species, multiple long-term experiments have demonstrated
11 transient growth increases (generally at deposition rates lower than 10 kg N/ha/yr) followed by
12 increased mortality, especially at higher rates of fertilization. In the western United States,
13 atmospheric nitrogen deposition has been shown to cause increased litter accumulation in the
14 soils and carbon storage in aboveground biomass, which, in turn, may lead to increased
15 susceptibility to more severe fires. Grassland communities that are adapted to low nutrient
16 supply can exhibit substantial sensitivity to nutrient enrichment effects of nitrogen deposition.
17 Invasive species of grass that may have been suppressed by nitrogen limitation can now better
18 compete and alter species dominance.

19 Two of the primary indicators of nitrogen overenrichment in forested watersheds are the
20 leaching of NO_3^- in soil drainage waters and the export of NO_3^- in stream water. Low carbon to
21 nitrogen (C:N) ratios in soils are also commonly related to increased nitrification, potential
22 increases in soil acidity, and releases of nitrate to receiving waters; however, these measurements
23 are not always widely available. Some of the highest nitrogen deposition has occurred in
24 Southern California, where researchers have documented measurable ecological changes related
25 to atmospheric deposition. Evidence from the two ecosystems discussed in this case study—
26 coastal sage scrub (CSS) and mixed conifer forest (MCF)—supports the finding that nitrogen
27 alters these habitats. In this case study, spatial information and observed, experimental effects
28 were used to help identify the trends in these ecosystems and describe the past and current spatial
29 extent of the ecosystems. Current analysis of the effects of terrestrial nutrient enrichment from
30 atmospheric nitrogen deposition in both CSS and MCF ecosystems seeks to improve scientific

1 understanding of the interactions among nitrogen deposition, fire events, and community
2 dynamics.

3 Due to data limitations, the assessment of ecological effects was based on a qualitative
4 weight-of-evidence approach based on the current scientific literature to determine benchmark
5 values for ecological effects due to nitrogen deposition in CSS and MCF communities. There are
6 sufficient data to relate an ecological effect to atmospheric nitrogen deposition.

7 For the CSS community, the following ecological thresholds were identified:

- 8 ■ 3.3 kg N/ha/yr – the amount of nitrogen uptake by a vigorous stand of CSS; above this
9 level, nitrogen may no longer be limiting
- 10 ■ 10 kg N/ha/yr – mycorrhizal community changes, CSS decline.

11 For the MCF community, the following ecological thresholds were identified:

- 12 ■ 3.1 kg N/ha/yr – shift from sensitive to tolerant lichen species
- 13 ■ 5.2 kg N/ha/yr – dominance of the tolerant lichen species
- 14 ■ 10.2 kg N/ha/yr – loss of sensitive lichen species
- 15 ■ 17 kg N/ha/yr – leaching of nitrate into streams.

16 **Table ES-3** displays the areas (in
17 ha) of CSS experiencing different total
18 nitrogen deposition levels.

19 Mutualistic fungal communities,
20 such as *arbuscular mycorrhizae* (AM),
21 increase the surface area and capacity for
22 nutrient uptake. However, in the presence of approximately 10 kg N/ha/yr, coarse AM
23 colonizations were depressed in number and volume.

24 Although CSS communities are fire resilient, nonnative grass seeds are quick to establish
25 in burned lands, reducing the water and nutrients available to CSS for reestablishment. With
26 increased fire frequencies and faster nonnative colonizations, CSS seed banks are eventually
27 eradicated from the soil, and the probability of reestablishment decreases significantly.

28 **Mixed Conifer Forest Case Study Area.** Measurements documenting increases in
29 atmospheric nitrogen deposition over MCF have been recorded with some regularity since the
30 1980s. The pressures exerted on MCF ecosystems in California form a gradient across the Sierra
31 Nevada Range and the San Bernardino Mountains. Nitrogen throughfall levels in the northern
32 Sierra Nevada Mountains are as low as 1.4 kg N/ha/yr, whereas forests in the western San

Table ES-3. Coastal Sage Scrub Ecosystem Area and Total Nitrogen Deposition

N Deposition (kg/ha/yr)	Area (hectares)	Percent of CSS Area, %
≥3.3	654,048	94
≥10	138,019	20

1 Bernardino Mountains experience measured throughfall nitrogen levels up to 33 to 71 kg
2 N/ha/yr.

3 **Table ES-4** shows the area of MCF experiencing levels of nitrogen deposition
4 corresponding to the identified benchmarks.

Table ES-4. Mixed Conifer Forest Ecosystem Area and Nitrogen Deposition

N Deposition (kg/ha/yr)	Area (hectares)	Percent of MCF Area, %
≥3.1	1,099,133	39
≥5.2	130,538	5
≥10.2	11,963	0.4
≥17	0	0

5
6 The proximity of CSS and MCF to population centers and recreational areas and the
7 potential value of these landscape types in providing regulating ecosystem services suggest that
8 the value of preserving CSS and MCF to California could be quite high. The primary cultural
9 ecosystem services associated with CSS and MCF are recreation, aesthetic, and non-use values.
10 Changes that might impact cultural ecosystem services in CSS resulting from nutrient
11 enrichment potentially include decline in CSS habitat, decline in protection of native species,
12 increase in abundance of nonnative grasses, and increase in wildfires. Additional ecosystem
13 services that might be impacted include fishing, hunting, wildlife viewing, and hiking. The
14 composition of species in CSS changes fire frequency and intensity because nonnative grasses
15 fuel more frequent and more intense wildfires. A healthy MCF ecosystem supports native
16 species, promotes water quality, and helps regulate fire intensity.

17 In MCF, maintaining water quality emerged as a regulating service that can be upset by
18 excessive nitrogen. When the soil becomes saturated, nitrates may leach into the surface water
19 and cause acidification. Additional nitrogen from MCF areas could further degrade waters that
20 are already stressed by numerous other sources of nutrients and pollution.

21 **ADDITIONAL EFFECTS**

22 Although this Risk and Exposure Assessment focused on acidification and nutrient
23 enrichment (from nitrogen), nitrogen and sulfur produce additional welfare effects, including
24 those related to visibility, climate, and material flows. Additional effects include the influence of

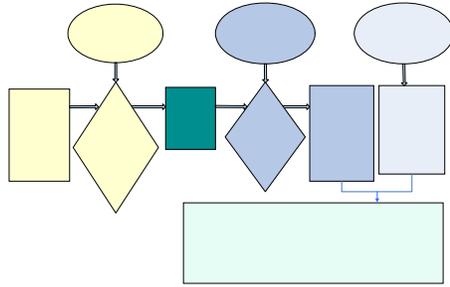
1 SO_x deposition on methylmercury production, N₂O effects on climate, nitrogen effects on
2 primary productivity and biogenic greenhouse gas fluxes, and phytotoxic effects on plants. While
3 these are important effects that are beyond the scope of this review, they were addressed
4 qualitatively. Impairment of visibility and materials damage also can result from atmospheric
5 particulate matter (PM), which is composed in part of sulfate- and nitrate-based particulates (i.e.,
6 ammonium sulfate and ammonium nitrate). These effects are being addressed in the PM NAAQS
7 review currently underway.

8 **CONCLUSIONS**

9 While there are many uncertainties associated with these analyses, from a scientific
10 perspective there is confidence that known or anticipated adverse ecological effects are occurring
11 under current ambient loadings of nitrogen and sulfur in sensitive ecosystems across the United
12 States. Of all the case study analyses, there is most confidence in the ecological responses,
13 effects, and benefits associated with aquatic acidification, and there is a fair amount of
14 confidence about those associated with terrestrial acidification. There is confidence in the
15 ecological responses, effects, and benefits associated with aquatic nitrogen nutrient enrichment
16 due to large contributions from nonatmospheric sources of nitrogen and the influence of both
17 oxidized and reduced forms of nitrogen, particularly in large watersheds and coastal areas.
18 However, there is a strong relationship between ecological responses and effects and
19 atmospheric deposition of nitrogen in high alpine lakes in the Rocky Mountains because
20 atmospheric deposition is the only source of nitrogen to these systems. In addition, there is
21 strong qualitative evidence regarding the relationships between ecological responses, effects, and
22 benefits attributable to terrestrial nitrogen nutrient enrichment; however, the relative
23 contributions of oxidized versus reduced forms of nitrogen must also be taken into account.
24 Based on the scientific analyses presented in the ISA and the Risk and Exposure Assessment,
25 negative ecological effects due to aquatic and terrestrial acidification may be the most useful in
26 terms of developing a secondary NO_x/SO_x NAAQS that reflects the ecological impacts due to
27 emissions of these pollutants.

REFERENCES

- 1
- 2 Baker, L.A., and P.L. Brezonik. 1988. Dynamic model of in-lake alkalinity generation. *Water*
3 *Resources Research* 24:65–74.
- 4 Baron, J.S. 2006. Hindcasting nitrogen deposition to determine ecological critical load.
5 *Ecological Applications* 16:433–439.
- 6 Lafrancois, B.M., K.R. Nydick, B.M. Johnson, and J.S. Baron. 2004. Cumulative effects of
7 nutrients and pH on the plankton of two mountain lakes. *Canadian Journal of Fisheries*
8 *and Aquatic Sciences* 61:1153–1165.
- 9 McKnight, D.M., R.L. Smith, J.P. Bradbury, J.S. Baron, and S. Spaulding. 1990. Phytoplankton
10 dynamics in three Rocky Mountain Lakes, Colorado U.S.A. *Arctic, Antarctic, and Alpine*
11 *Research* 22:264–274.
- 12 Saros, J.E., S.J. Interlandi, A.P. Wolfe, and D.R. Engstrom. 2003. Recent changes in the diatom
13 community structure of lakes in the Beartooth Mountain Range, U.S.A. *Arctic, Antarctic,*
14 *and Alpine Research* 35:18–23.
- 15 Saros, J.E., T.J. Michel, S.J. Interlandi, and A.P. Wolfe. 2005. Resource requirements of
16 *Asterionella formosa* and *Fragilaria crotonensis* in oligotrophic alpine lakes:
17 implications for recent phytoplankton community reorganizations. *Canadian Journal of*
18 *Fisheries and Aquatic Sciences* 62:1681–1689.
- 19 Sullivan, T.J., C.T. Driscoll, B.J. Cosby, I.J. Fernandez, A.T. Herlihy, J. Zhai, R. Stemberger,
20 K.U. Snyder, J.W. Sutherland, S.A. Nierzwicki-Bauer, C.W. Boylen, T.C. McDonnell,
21 and N.A. Nowicki. 2006. *Assessment of the Extent to Which Intensively-Studied Lakes*
22 *are Representative of the Adirondack Mountain Region*. Final report. New York State
23 Energy Research and Development Authority (NYSERDA), Albany, NY. Available at
24 <http://nysl.nysed.gov/uhtbin/cgisirsi/Qcwd6NzFby/NYSL/138650099/8/4298474>
25 (accessed November 1, 2007).
- 26 Sverdrup, H., and P. Warfvinge. 1993. *The effect of soil acidification on the growth of trees,*
27 *grass and herbs as expressed by the (Ca+ Mg+ K)/Al ratio*. Reports in Ecology and
28 Environmental Engineering 2. Lund University, Department of Chemical Engineering,
29 Lund, Sweden.
- 30 U.S. EPA (Environmental Protection Agency). 2006. *Ecological Benefits Assessment Strategic*
31 *Plan*. EPA-240-R-06-001. U.S. Environmental Protection Agency, Office of the
32 Administrator. Washington, DC. Available at <http://www.epa.gov/economics>.
- 33 U.S. EPA (Environmental Protection Agency). 2008. *Integrated Science Assessment (ISA) for*
34 *Oxides of Nitrogen and Sulfur—Ecological Criteria (Final Report)*. EPA/600/R-
35 08/082F. U.S. Environmental Protection Agency, National Center for Environmental
36 Assessment—RTP Division, Office of Research and Development, Research Triangle
37 Park, NC. Available at <http://cfpub.epa.gov/ncea/cfm/recordisplay.cfm?deid=201485>.

1
2

3

1.0 INTRODUCTION

1.1 RATIONALE AND BACKGROUND FOR JOINT REVIEW

4 The U.S. Environmental Protection Agency (EPA or the Agency) is conducting a joint
5 review of the existing secondary (welfare-based) National Ambient Air Quality Standards
6 (NAAQS) for nitrogen oxides (NO_x) and sulfur oxides (SO_x), which are currently defined in
7 terms of nitrogen dioxide (NO₂) and sulfur dioxide (SO₂), respectively.¹ Sections 108 and 109 of
8 the Clean Air Act (CAA or the Act) govern the establishment and periodic review of the
9 NAAQS and of the air quality criteria upon which the standards are based. The NAAQS are
10 established for pollutants that may reasonably be anticipated to endanger public health or welfare
11 and whose presence in the ambient air results from numerous or diverse mobile or stationary
12 sources. The NAAQS are based on air quality criteria that reflect the latest scientific knowledge,
13 which is useful in indicating the kind and extent of identifiable effects on public health or
14 welfare that may be expected from the presence of the pollutant in ambient air. Based on
15 periodic reviews of the air quality criteria and standards, EPA makes revisions to the criteria and
16 standards and promulgates any new standards as may be appropriate. The Act also requires that
17 an independent scientific review committee advise the Administrator as part of this NAAQS
18 review process, a function now performed by the Clean Air Scientific Advisory Committee
19 (CASAC).
20

21 In conducting this periodic review of the NO₂ and SO₂ secondary NAAQS, EPA has
22 decided to jointly assess the scientific information, associated risks, and standards relevant to
23 protecting the public welfare from adverse effects associated with NO_x and SO_x. As noted in
24 Section 1.2 of this report, EPA has historically defined the NAAQS for these pollutants in terms

¹ EPA is also conducting independent reviews of the primary (health-based) NAAQS for NO_x and SO_x.

1 of the specific compounds NO₂ and SO₂, which serve as indicators of the broader set of
2 compounds that comprise NO_x and SO_x, respectively. The species of nitrogen and sulfur
3 compounds and the types of related ecological effects that are being considered within the scope
4 of this review are discussed in Section 1.3 of this report. A joint secondary review of these
5 pollutants is being conducted because NO_x, SO_x, and their associated transformation products are
6 linked from an atmospheric chemistry perspective, as well as from an environmental effects
7 perspective, and because the National Research Council (NRC) has recommended that EPA
8 consider multiple pollutants, as appropriate, in forming the scientific basis for the NAAQS
9 (NRC, 2004). This is the first time since the NAAQS were established in 1971 that a joint review
10 of these two pollutants has been conducted. There is a strong basis for considering these
11 pollutants together at this time, building upon EPA's and CASAC's past recognition of the
12 interactions of these pollutants and on the growing body of scientific information that is now
13 available related to these interactions and associated ecological effects. A series of policy-
14 relevant questions that help to frame this review are presented in Section 1.4 of this report,
15 together with an overview of how secondary NAAQS for NO_x and SO_x might be structured to
16 reflect the complex interactions among relevant species of these pollutants in an ecologically
17 meaningful way. As discussed in the CAA [Section 109(b)(2)], the purpose of a secondary
18 NAAQS is to protect the public welfare from any known or anticipated adverse effects
19 associated with the presence of such air pollutants in the ambient air.

20 This joint review is organized according to EPA's current NAAQS review process, which
21 consists of four major components and related documents: an *Integrated Review Plan* (U.S. EPA,
22 2007), the *Integrated Science Assessment (ISA) for Oxides of Nitrogen and Sulfur—Ecological*
23 *Criteria (Final Report)*(ISA) (U.S. EPA, 2008), the Risk and Exposure Assessment, and a policy
24 assessment and rulemaking notices. The *Integrated Review Plan* provides the framework and
25 schedule for this review and identifies policy-relevant questions to be addressed in the other
26 components of the review. The ISA, released on December 12, 2008, provides an integrative
27 assessment of the relevant scientific information and forms the scientific basis for the
28 assessments presented in this Risk and Exposure Assessment, which describes the progress to
29 date on the assessments being conducted as part of the third component of the review process. To
30 view related documents developed as part of the planning and science assessment phases of this

1 review (e.g., *Integrated Review Plan*, the ISA), see
2 <http://www.epa.gov/ttn/naaqs/standards/no2so2sec/index.html>.

3 When complete, the Risk and Exposure Assessment will evaluate the exposures of
4 ecological receptors to both ambient and deposited species of NO_x and SO_x, as well as their
5 transformation products (including reduced forms of ambient nitrogen), and assess, both
6 quantitatively and qualitatively, the risks associated with these exposures. Where possible, the
7 contributions of various sources and forms of atmospheric nitrogen to these risks are
8 characterized. The following bullets outline the organization of this final draft report, which, to
9 the degree possible, reflects the components of the Risk and Exposure Assessment:

- 10 ▪ **Chapter 1** provides an overview of this review; a history of past reviews and other
11 relevant scientific assessments and EPA actions; a discussion of the scope of this joint
12 NO_x and SO_x review; and a series of policy-relevant questions, together with an
13 overview of how secondary NAAQS for NO_x and SO_x might be structured.
- 14 ▪ **Chapter 2** provides an overview of the Risk and Exposure Assessment, including the
15 scope and approach to assessing current conditions for a targeted effect, a summary of the
16 case study areas, a discussion of the identification and selection of ecosystem services,
17 and a discussion on addressing uncertainty throughout the review.
- 18 ▪ **Chapter 3** addresses the relevant air quality issues associated with this review, including
19 the sources, emissions, and deposition of total reactive nitrogen and sulfur and their
20 current contributions to ambient conditions. Both spatial and temporal characterizations
21 of ambient concentrations of nitrogen and sulfur and the contributions of ambient
22 concentrations of nitrogen and sulfur to deposition are explored in select case study areas.
23 In addition, there is a discussion on the relationship between atmospheric concentrations
24 and deposition and how the Atmospheric Deposition Transformation Function might be
25 structured (see **Figure 1.4-1**).
- 26 ▪ **Chapter 4** focuses on acidification, with an overview of the relevant science and
27 progress on case study analyses and developing the associated ecological effect functions
28 (see **Figure 1.4-1**) for both aquatic and terrestrial acidification.
- 29 ▪ **Chapter 5** focuses on nitrogen nutrient enrichment, with an overview of the relevant
30 science and progress on case study analyses and developing the associated ecological

1 effect functions (see **Figure 1.4-1**) for both aquatic and terrestrial nitrogen nutrient
2 enrichment (commonly referred to as nutrient enrichment).

- 3 ■ **Chapter 6** qualitatively addresses additional effects, including visibility, climate, and
4 materials. There is a discussion on the interactions between sulfur and methylmercury
5 production, nitrous oxide (N₂O) effects on climate, nitrogen addition effects on primary
6 productivity and biogenic greenhouse gas fluxes, and phytotoxic effects on plants.
- 7 ■ **Chapter 7** characterizes adversity from a scientific perspective and synthesizes the case
8 study results from Chapters 3 through 5. Chapter 7 also summarizes how the key findings
9 of the Risk and Exposure Assessment will be carried into the policy assessment portion
10 of this review.

11 **1.2 HISTORY**

12 **1.2.1 History of the Secondary NO₂ NAAQS**

13 On April 30, 1971, EPA promulgated identical primary and secondary NAAQS for NO₂
14 under Section 109 of the CAA. The standards were set at 0.053 parts per million (ppm), annual
15 average (36 FR 8186). In 1982, EPA published the air quality criteria document (AQCD) *Air*
16 *Quality Criteria for Oxides of Nitrogen* (U.S. EPA, 1982), which updated the scientific criteria
17 for NO_x, upon which the initial NO₂ standards were based. On February 23, 1984, EPA proposed
18 to retain these standards (49 FR 6866). After taking into account public comments, EPA
19 published the final decision to retain these standards on June 19, 1985 (50 FR 25532).

20 On July 22, 1987, EPA announced that it was undertaking plans to revise the 1982 NO_x
21 AQCD (52 FR 27580), and in November 1991, EPA released an updated draft AQCD for
22 CASAC and public review and comment (56 FR 59285). This draft document provided a
23 comprehensive assessment of the available scientific and technical information on health and
24 welfare effects associated with NO₂ and other NO_x. CASAC reviewed the draft document at a
25 meeting held on July 1, 1993, and concluded in a closure letter to the Administrator that the
26 document “provides a scientifically balanced and defensible summary of current knowledge of
27 the effects of this pollutant and provides an adequate basis for EPA to make a decision as to the
28 appropriate NAAQS for NO₂” (Wolff, 1993). The AQCD *Air Quality Criteria for Oxides of*
29 *Nitrogen* was then finalized (U.S. EPA, 1993).

1 EPA also prepared a Staff Paper that summarized an air quality assessment for NO₂
2 conducted by the Agency (McCurdy, 1994). This Staff Paper summarized and integrated the key
3 studies and scientific evidence contained in the revised NO_x AQCD and identified the critical
4 elements to be considered in the review of the NO₂ NAAQS. CASAC reviewed two drafts of the
5 Staff Paper and concluded in a closure letter to the Administrator that the document provided a
6 “scientifically adequate basis for regulatory decisions on nitrogen dioxide” (Wolff, 1995). In
7 September 1995, EPA finalized the Staff Paper, entitled *Review of the National Ambient Air*
8 *Quality Standards for Nitrogen Dioxide: Assessment of Scientific and Technical Information*
9 (U.S. EPA, 1995a).

10 In October 1995, the Administrator announced her proposed decision not to revise either
11 the primary or secondary NAAQS for NO₂ (60 FR 52874; October 11, 1995). A year later, the
12 Administrator made a final determination not to revise the NAAQS for NO₂ after careful
13 evaluation of the comments received on the proposal (61 FR 52852; October 8, 1996). The level
14 for both the existing primary and secondary NAAQS for NO₂ is 0.053 ppm (100 micrograms per
15 cubic meter [$\mu\text{g}/\text{m}^3$] of air), annual arithmetic average, calculated as the arithmetic mean of the
16 1-hour NO₂ concentrations.

17 **1.2.2 History of the Secondary SO₂ NAAQS**

18 Based on the 1970 AQCD *Air Quality Criteria for Sulfur Oxides* (DHEW, 1970), EPA
19 promulgated primary and secondary NAAQS for SO₂ under Section 109 of the CAA on April 30,
20 1971 (36 FR 8186). The secondary standards included a standard at 0.02 ppm in an annual
21 arithmetic mean and a 3-hour average of 0.5 ppm, not to be exceeded more than once per year.
22 These secondary standards were established solely on the basis of vegetation-effects evidence. In
23 1973, revisions made to Chapter 5 (*Effects of Sulfur Oxide in the Atmosphere on Vegetation*) of
24 the AQCD *Effects of Sulfur Oxides in the Atmosphere on Vegetation; Revised Chapter 5 for Air*
25 *Quality Criteria for Sulfur Oxides* (U.S. EPA, 1973) indicated that it could not properly be
26 concluded that the vegetation injury reported resulted from the average SO₂ exposure over the
27 growing season, rather than from short-term peak concentrations. Therefore, EPA proposed 38
28 FR 11355 and then finalized 38 FR 25678, a revocation of the annual mean secondary standard.
29 At that time, EPA was aware that SO_x have other public welfare effects, including effects on
30 materials, visibility, soils, and water; however, the available data were considered insufficient to

1 establish a quantitative relationship between specific SO_x concentrations and effects needed for
2 setting a standard (38 FR 25679).

3 In 1979, EPA announced that it was revising the 1973 SO_x AQCD concurrently with that
4 for particulate matter (PM) and would produce a combined PM and SO_x criteria document.
5 Following its review of a draft revised criteria document in August 1980, CASAC concluded that
6 acidifying deposition was a topic of extreme scientific complexity because of the difficulty in
7 establishing firm quantitative relationships among (1) emissions of relevant pollutants (e.g., SO₂,
8 NO_x), (2) formation of acidifying wet and dry deposition products, and (3) effects on terrestrial
9 and aquatic ecosystems. CASAC also noted that acidifying deposition involves, at a minimum,
10 several different criteria pollutants: SO_x, NO_x, and the fine particulate fraction of suspended
11 particles. CASAC felt that any document on this subject should address both wet and dry
12 deposition because dry deposition was believed to account for at least one-half of the total
13 acidifying deposition problem.

14 For these reasons, CASAC recommended that a separate, comprehensive document on
15 acidifying deposition be prepared prior to any consideration of using the NAAQS as a regulatory
16 mechanism for the control of acidifying deposition. CASAC also suggested that a discussion of
17 acidifying deposition be included in the AQCD for NO_x, PM, and SO_x. Following CASAC
18 closure on the criteria document for SO₂ in December 1981, EPA's Office of Air Quality
19 Planning and Standards (OAQPS) published a Staff Paper in November 1982, but the paper did
20 not directly assess the issue of acidifying deposition. Instead, EPA subsequently prepared the
21 following documents: *The Acidic Deposition Phenomenon and Its Effects: Critical Assessment*
22 *Review Papers, Volumes I and II* (U.S. EPA, 1984a, b) and *The Acidic Deposition Phenomenon*
23 *and Its Effects: Critical Assessment Document* (U.S. EPA, 1985) (53 FR 14935-14936). Though
24 these documents were not considered criteria documents and did not undergo CASAC review,
25 they represented the most comprehensive summary of relevant scientific information completed
26 by EPA to that point.

27 On April 26, 1988 (53 FR 14926), EPA proposed not to revise the existing primary and
28 secondary standards. This proposal regarding the secondary SO₂ NAAQS was due to the
29 Administrator's conclusions that (1) based upon the then-current scientific understanding of the
30 acidifying deposition problem, it would be premature and unwise to prescribe any regulatory
31 control program at that time, and (2) when the fundamental scientific uncertainties had been

1 reduced through ongoing research efforts, EPA would draft and support an appropriate set of
2 control measures.

3 **1.2.3 History of Related Assessments and Agency Actions**

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

10 In spite of the complexities and significant remaining uncertainties associated with the
11 acidifying deposition problem, it soon became clear that a program to address acidifying
12 deposition was needed. The Amendments to the CAA passed by Congress and signed into law by
13 the president on November 15, 1990, included numerous separate provisions related to the
14 acidifying deposition problem that reflect the comprehensive approach envisioned by Congress.
15 The primary and most important of the provisions, Title IV of the CAA Amendments,
16 established the Acid Rain Program to reduce SO₂ emissions by 10 million tons and NO_x
17 emissions by 2 million tons from 1980 emission levels to achieve reductions over broad
18 geographic regions. In this provision, Congress included a statement of findings that led them to
19 take action, concluding that (1) the presence of acid compounds and their precursors in the
20 atmosphere and in deposition from the atmosphere represents a threat to natural resources,
21 ecosystems, materials, visibility, and public health; (2) the problem of acidifying deposition is of
22 national and international significance; and (3) current and future generations of Americans will
23 be adversely affected by delaying measures to remedy the problem.

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

27 Third, Congress—clearly envisioning that further action might be necessary in the long
28 term to address any problems remaining after implementation of the Title IV program and
29 reserving judgment on the form that action could take—included Section 404 of the 1990
30 Amendments (CAA Amendments of 1990, Pub. L. 101-549, § 404), requiring EPA to conduct a

1 study on the feasibility and effectiveness of an acidifying deposition standard or standards to
2 protect “sensitive and critically sensitive aquatic and terrestrial resources.” At the conclusion of
3 the study, EPA was to submit a report to Congress. Five years later, in fulfillment of this
4 requirement, EPA submitted its report, entitled *Acid Deposition Standard Feasibility Study:
5 Report to Congress* (U.S. EPA, 1995b). The report concluded that establishing acidifying
6 deposition standards for sulfur and nitrogen deposition may at some point in the future be
7 technically feasible, although appropriate deposition loads for these acidifying chemicals could
8 not be defined with reasonable certainty at that time.

9 Fourth, the 1990 Amendments also added new language to sections of the CAA
10 pertaining to the scope and application of the secondary NAAQS designed to protect the public
11 welfare. Specifically, the definition of “public welfare” in Section 302(h) was expanded to state
12 that the welfare effects identified should be protected from adverse effects associated with
13 criteria air pollutants “...whether caused by transformation, conversion, or combination with
14 other air pollutants.” This change has particular relevance to this review because the
15 transformation products of NO_x and SO_x are associated with environmental impacts.

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

30 In a related matter, the Office of the Secretary of the U.S. Department of Interior (DOI)
31 requested in 2000 that EPA initiate a rulemaking proceeding to enhance the air quality in

1 national parks and wilderness areas to protect resources and values that are being adversely
2 affected by air pollution. Included among the effects of concern identified in the request were the
3 acidification of streams, surface waters, and/or soils; eutrophication of coastal waters;
4 impairment of visibility; and foliar injury from ozone.

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

9 The 2005 NAPAP report states that "... scientific studies indicate that the emission
10 reductions achieved by Title IV are not sufficient to allow recovery of acid-sensitive ecosystems.
11 Estimates from the literature of the scope of additional emission reductions that are necessary in
12 order to protect acid-sensitive ecosystems range from approximately 40-80% beyond full
13 implementation of Title IV.... The results of the modeling presented in this Report to Congress
14 indicate that broader recovery is not predicted without additional emission reductions" (NAPAP,
15 2005).

16 Given the state of the science as described in the ISA and in other recent reports, such as
17 the 2005 NAPAP report, EPA believes it is appropriate, in the context of evaluating the
18 adequacy of the current NO₂ and SO₂ secondary standards in this review, to revisit the question
19 of the appropriateness and the feasibility of setting a secondary NAAQS to address remaining
20 known or anticipated adverse public welfare effects resulting from the acidifying and nutrient
21 deposition of these criteria pollutants and their transformation products. This document
22 comprises the Risk and Exposure Assessment portion of the review.

23 **1.3 SCOPE OF THE RISK AND EXPOSURE ASSESSMENT FOR THE** 24 **CURRENT REVIEW**

25 **1.3.1 Species of Nitrogen Included in the Analyses**

26 The sum of mono-nitrogen oxides—nitrogen dioxide (NO₂) and nitric oxide (NO)—
27 typically are referred to as nitrogen oxides (NO_x) in the atmospheric science community. More
28 formally, the family of NO_x includes any gaseous combination of nitrogen and oxygen (e.g.,
29 NO₂, NO, N₂O, nitrogen trioxide [N₂O₃], nitrogen tetroxide [N₂O₄], and dinitrogen pentoxide
30 [N₂O₅]).

1 With regard to NO_x , it is also necessary in this review to distinguish between the
2 definition of “nitrogen oxides” as it appears in the enabling legislation related to the NAAQS and
3 the definition commonly used in the air pollution research and management community. In this
4 document, the term “oxides of nitrogen” and “nitrogen oxides” refer to all forms of oxidized
5 nitrogen compounds, including NO , NO_2 , and all other oxidized nitrogen-containing compounds
6 transformed from NO and NO_2 . This definition is supported by Section 108(c) of the CAA,
7 which states that “Such criteria [for oxides of nitrogen] shall include a discussion of nitric and
8 nitrous acids, nitrites, nitrates, nitrosamines, and other carcinogenic and potentially carcinogenic
9 derivatives of oxides of nitrogen.” The term used by the scientific community to represent the
10 complete set of oxidized nitrogen compounds, including those listed in CAA Section 108(c), is
11 total oxidized nitrogen (NO_y). NO_y includes all nitrogen oxides, including gaseous nitrate species
12 such as nitric acid (HNO_3) and peroxyacyl nitrates (PAN).

13 In addition to oxidized forms of nitrogen, reduced forms of nitrogen also contribute to the
14 atmospheric chemistry that leads to the deposition of ambient nitrogen species to the
15 environment. Reduced atmospheric nitrogen species include ammonia (NH_3) and ammonium ion
16 (NH_4^+), the sum of which is referred to as reduced nitrogen (NH_x). Total reactive nitrogen is
17 recognized as the combination of both oxidized and reduced forms of nitrogen that are
18 biologically available (i.e., forms other than the stable form of gaseous nitrogen [N_2]).
19 Atmospheric nitrogen deposition often is delineated further as dry (e.g., gas and particulate
20 phases) or as wet (e.g., precipitation-derived ion phase) (see **Figure 1.3-1**).

21 In many areas, multiple forms of nitrogen from a variety of atmospheric and other
22 sources enter ecosystems. The scientific community has long recognized that the effects from
23 atmospheric deposition of nitrogen to ecosystems are due to both oxidized and reduced forms,
24 rather than to one form alone. As a result, much of the published research on ecological response
25 to nitrogen does not differentiate between the various sources of nitrogen, but instead reports
26 only total nitrogen inputs to the ecosystem.

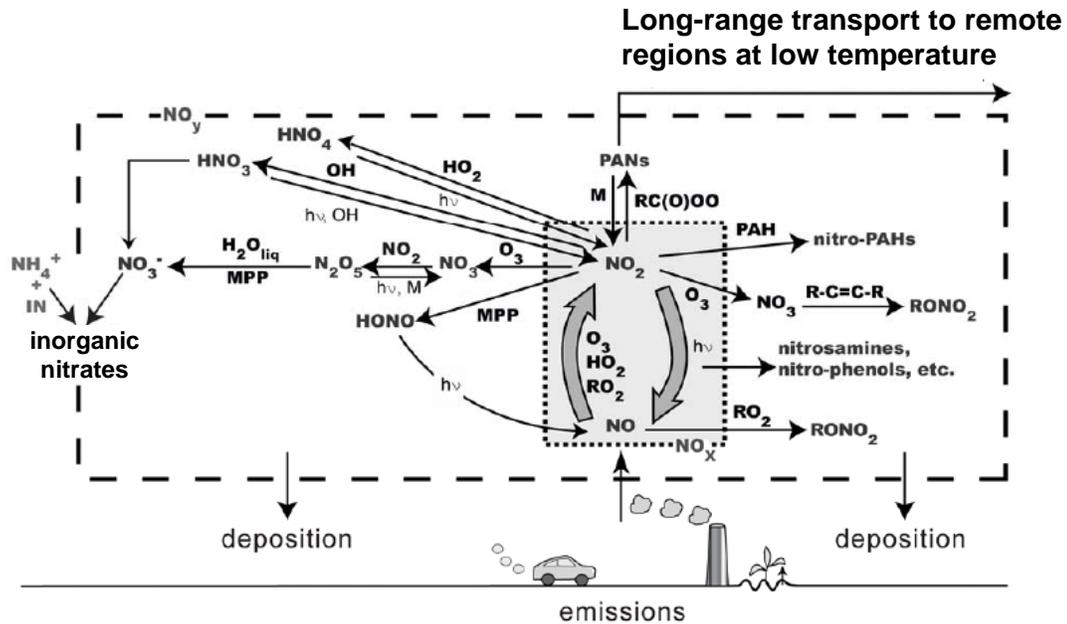
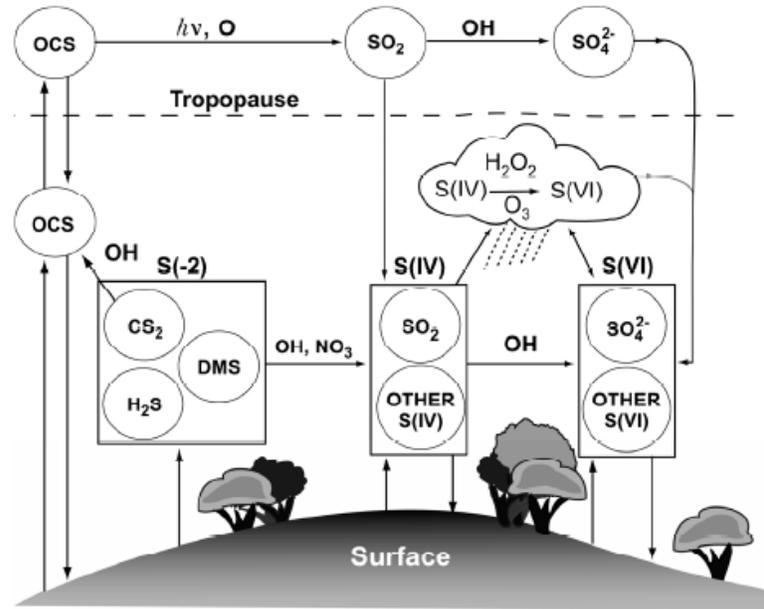


Figure 1.3-1. Schematic diagram of the cycle of reactive, oxidized nitrogen species in the atmosphere. Particulate-phase organic nitrates are also formed from the species on the right side of the figure (U.S. EPA, 2008).

Note: IN = inorganic particulate species (e.g., sodium [Na⁺], calcium [Ca²⁺]), MPP = multiphase processes, PAN = peroxyacetyl nitrates, PAH = polycyclic aromatic hydrocarbon, $h\nu$ = a solar photon, R = an organic radical.

1.3.2 Species of Sulfur Included in the Analyses

SO₂ is one of a group of substances known as “oxides of sulfur”, or SO_x, which include multiple gaseous species (e.g., SO₂, sulfur monoxide [SO], sulfur trioxide [SO₃], thiosulfate [S₂O₃], sulfur heptoxide [S₂O₇]) and particulates (e.g., ammonium sulfate [(NH₄)₂SO₄]) (Figure 1.3-2). Acidification can result from the atmospheric deposition of SO_x and NO_x; in acidifying deposition, these species combine with water in the atmosphere to form sulfuric acid (H₂SO₄) and HNO₃. Due to known acute effects on plants, SO₂ served as the chemical indicator for SO_x species in previous NAAQS reviews.



1
2 **Figure 1.3-2.** Schematic diagram of the cycle of sulfur species in the atmosphere
3 (adapted from Berresheim et al. (1995); used with permission.) .

4 **Note:** OCS = carbonyl sulfide, DMS = dimethyl sulfide, S(IV) = S^{+4} , S(VI) = S^{+6} .

5 1.3.3 Overview of Nitrogen- and Sulfur-Related Ecological Effects

6 The ecological effects of nitrogen and sulfur are caused both by the gas-phase and
7 atmospheric deposition of the pollutants. The current secondary NAAQS were set to protect
8 against direct damage to vegetation by exposure to gas-phase NO_x or SO_x . Acute and chronic
9 exposures to SO_2 can have phytotoxic effects on vegetation, such as foliar injury, decreased
10 photosynthesis, and decreased growth. Similarly, exposure to sufficient concentrations of NO_2 ,
11 NO , PAN, and HNO_3 can cause foliar injury, decreased photosynthesis, and decreased growth
12 (U.S. EPA 2008).

13 With respect to direct gas-phase effects, the ISA for the secondary NAAQS review
14 determined the following:

15 *The evidence is sufficient to infer a causal relationship between exposure to SO_2 , NO ,*
16 *NO_2 , PAN, and HNO_3 and injury to vegetation.*

17 Even though these gas-phase chemicals will cause phytotoxicity, the evidence indicates
18 there is little new evidence that current concentrations of gas-phase sulfur or nitrogen oxides are
19 not high enough to cause phytotoxic effects. One exception is that some studies indicate that

1 current HNO₃ concentrations may be contributing to the decline of lichen species in the Los
2 Angeles basin. (U.S. EPA, 2008).

3 Deposition of nitrogen-containing and sulfur-containing compounds that are derived from
4 NO_x and SO_x may be wet (e.g., rain and snow), occult (e.g., cloud and fog), and dry (e.g., gases
5 and particles) and can affect ecosystem biogeochemistry, structure, and function. Nitrogen and
6 sulfur interactions in the environment are highly complex. Both are essential, and sometimes
7 limiting, nutrients needed for growth and productivity. Excess nitrogen (both oxidized and
8 reduced forms) or sulfur can lead to acidification, nitrogen nutrient enrichment, eutrophication,
9 and sulfur-mediated mercury methylation. Acidification causes a cascade of effects that alter
10 both terrestrial and aquatic ecosystems. These effects include slower growth, the injury or death
11 of forest vegetation, and the localized extinction of fish and other aquatic species.

12 With respect to acidification, the ISA determined the following:

13 *The evidence is sufficient to infer a causal relationship between acidifying deposition and*
14 *effects on*

15 *(1) biogeochemistry related to terrestrial and aquatic ecosystems;*

16 *(2) biota in terrestrial and aquatic ecosystems.*

17 The ISA highlights evidence from two well-studied areas to provide more detail on how
18 acidification affects ecosystems: the Adirondack Case Study Area (New York) and the
19 Shenandoah Case Study Area (Virginia) (U.S., EPA, 2008, Section 3.2). In the Adirondack Case
20 Study Area, the current rates of nitrogen and sulfur deposition exceed the amount that would
21 allow recovery of the most acid-sensitive lakes. In the Shenandoah Case Study Area, legacy
22 sulfate has accumulated in the soil and is slowly released from the soil into stream water, where
23 it causes acidification and makes this region sensitive to current loading. Models suggest that the
24 number of acidic streams will increase under the current deposition rates (U.S. EPA, 2008,
25 Section 3.2). The ISA highlights forests in the Adirondack Case Study Area of New York, Green
26 Mountains of Vermont, White Mountains of New Hampshire, and the Allegheny Plateau of
27 Pennsylvania, and high-elevation forest ecosystems in the southern Appalachians as the regions
28 most sensitive to terrestrial acidification effects from acidifying deposition (U.S. EPA, 2008,
29 Section 3.2). In this Risk and Exposure Assessment, these areas are targeted for the air quality
30 modeling presented in Chapter 3 and the case study analyses presented in Chapter 4 of this
31 report.

1 In addition to acidification, NO_x acts with other forms of total reactive nitrogen
2 (including reduced nitrogen) to increase the total amount of available nitrogen in ecosystems.
3 The contribution of nitrogen deposition to total nitrogen load varies among ecosystems.
4 Atmospheric nitrogen deposition is the main source of new nitrogen to most headwater streams,
5 high-elevation lakes, and low-order streams. Atmospheric nitrogen deposition contributes to the
6 total nitrogen load in terrestrial, wetland, freshwater, and estuarine ecosystems that receive
7 nitrogen through multiple pathways (i.e., biological nitrogen-fixation, agricultural land runoff,
8 and wastewater effluent discharges) (U.S. EPA, 2008, Section 3.3). Nitrogen deposition alters
9 numerous biogeochemical indicators, including primary productivity that leads to changes in
10 community composition and eutrophication.

11 With respect to nitrogen nutrient enrichment, the ISA determined the following:

12 *The evidence is sufficient to infer a causal relationship between nitrogen deposition, to*
13 *which NO_x and NH_x contribute, and the alteration of the following:*

14 (1) *Biogeochemical cycling of nitrogen and carbon in terrestrial, wetland, freshwater*
15 *aquatic, and coastal marine ecosystems*

16 (2) *Biogenic flux of methane and nitrous oxide in terrestrial and wetland ecosystems*

17 (3) *Species richness, species composition, and biodiversity in terrestrial, wetland,*
18 *freshwater aquatic, and coastal marine ecosystems.*

19 In aquatic ecosystems, wet deposition loads of approximately 1.5 to 2 kg N/ha/yr are
20 reported to cause alterations in diatom communities of freshwater lakes and to impair water
21 quality in the western United States (U.S. EPA, 2008, Section 3.3). In estuarine ecosystems,
22 additional nitrogen from anthropogenic atmospheric sources contributes to the total nitrogen
23 loading and to increased phytoplankton and algal productivity, which leads to eutrophication.
24 Estuary eutrophication is a detrimental ecological problem indicated by water quality
25 deterioration, resulting in numerous adverse effects, including hypoxic zones, species mortality,
26 and harmful algal blooms. The ISA indicates that the contribution of atmospheric deposition to
27 total nitrogen loads can be >40% in highly eutrophic estuaries. The Chesapeake Bay is an
28 example of a large, well-studied estuary that receives as much as 30% of its total nitrogen load
29 from the atmosphere (U.S. EPA, 2008, Section 3.3).

30 In terrestrial ecosystems, there are multiple chemical indicators for the alteration of the
31 biogeochemical cycling of nitrogen that is caused by total reactive nitrogen deposition. Nitrate

1 leaching is a well-documented effect that indicates the ecosystem is receiving more nitrogen than
2 it uses; the onset of leaching is calculated to be between 8 and 10 kg/ha/yr for eastern forests
3 (U.S. EPA, 2008, Section 3.3). Nitrogen deposition can cause ecological effects prior to the onset
4 of nitrate leaching. For example, nitrogen deposition affects primary productivity, thereby
5 altering terrestrial carbon cycling. This may result in shifts in population dynamics, species
6 composition, community structure, and, in extreme instances, ecosystem type. Lichen are the
7 most sensitive terrestrial taxa, with documented adverse effects occurring at 3 kg N/ha/yr
8 (Pacific Northwest and Southern California); 5 kg N/ha/yr correlates to the onset of declining
9 biodiversity within grasslands (Minnesota and the European Union); and 10 kg N/ha/yr causes
10 changes in community composition of Alpine ecosystems and forest encroachment into
11 temperate grasslands (U.S. EPA, 2008, Section 3.3). Some of the aquatic and terrestrial
12 ecosystems highlighted in the ISA are targeted for the air quality modeling presented in Chapter
13 3 and the case study analyses presented in Chapter 5 of this report.

14 There is increasing evidence on the relationship between sulfur deposition and increased
15 methylation of mercury in aquatic environments; this effect occurs only where other factors are
16 present at levels within a range to allow methylation. The production of methylmercury requires
17 the presence of sulfate and mercury, but the amount of methylmercury produced varies with
18 oxygen content, temperature, pH, and supply of labile organic carbon (U.S. EPA, 2008, Section
19 3.4). In watersheds where changes in sulfate deposition did not produced an effect, one or several
20 of those interacting factors were not in the range required for meaningful methylation to occur
21 (U.S. EPA, 2008, Section 3.4). Watersheds with conditions known to be conducive to mercury
22 methylation can be found in the northeastern United States and southeastern Canada. The
23 relationship between sulfur and methylmercury production is addressed qualitatively in Chapter
24 6 of this report.

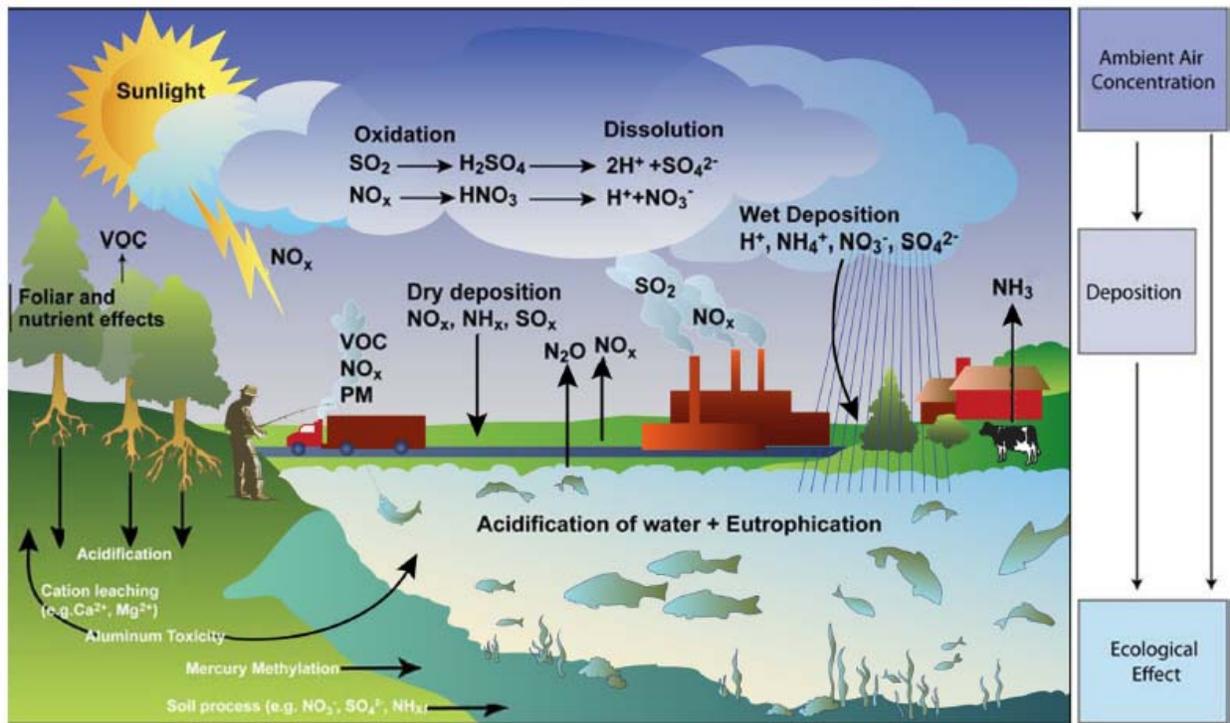
25 With respect to sulfur deposition and mercury methylation, the ISA determined the
26 following:

27 *The evidence is sufficient to infer a causal relationship between sulfur deposition and*
28 *increased mercury methylation in wetlands and aquatic environments.*

29 In terrestrial and wetland ecosystems, total reactive nitrogen deposition alters biogenic
30 sources and sinks of N₂O and methane—two potent greenhouse gases—resulting in a higher
31 emission to the atmosphere of these gases. Terrestrial soil is the largest source of N₂O,

1 accounting for 60% of global emissions. Total reactive nitrogen deposition increases the flux of
 2 N_2O in coniferous forests, deciduous forests, grasslands, and wetlands. Nitrogen deposition
 3 significantly reduces methane uptake in coniferous and deciduous forests, with a reduction of
 4 28% and 45%, respectively. In wetlands, nitrogen addition increases methane production, but has
 5 no significant effect on methane uptake (U.S. EPA, 2008, Section 3.4). These effects are also
 6 addressed qualitatively in Chapter 6 of this report.

7 A summary illustration of NO_x and SO_x effects on the environment is presented in
 8 **Figure 1.3-3.**



9
 10 **Figure 1.3-3.** Nitrogen and sulfur cycling and interactions in the environment.

11 **Note:** VOC = volatile organic compound.

12 1.4 POLICY-RELEVANT QUESTIONS

13 As many years of research have clearly demonstrated, the ecological effects associated
 14 with acidification and nutrient enrichment derive from both oxidized and reduced nitrogen, not
 15 NO_x alone, which is the current listed criteria pollutant. The policy-relevant questions driving
 16 this review recognize that the effects of NO_x occur as part of the overall effects of total reactive
 17 nitrogen and address the need to understand the role of NO_x relative to other sources of reactive
 18 nitrogen that contribute to adverse public welfare effects. Throughout the ISA and the Risk and

1 Exposure Assessment, public welfare effects due to total reactive nitrogen are examined, and
2 where possible, the contributions to these effects from oxidized and reduced forms of nitrogen
3 are assessed. For this secondary NO_x/SO_x NAAQS review, the main policy-relevant questions
4 include the following:

- 5 ▪ What are the known or anticipated welfare effects influenced by ambient NO_x, an
6 important component of total reactive nitrogen, and SO_x, and for which effects is there
7 sufficient information available to be useful as a basis for considering distinct secondary
8 standard(s)?
- 9 ▪ What is the nature and magnitude of ecosystem responses to total reactive nitrogen, of
10 which NO_x contributes, and SO_x that are understood to have known or anticipated
11 detrimental public welfare effects, and what is the variability associated with those
12 responses (including ecosystem type, climatic conditions, interactions with other
13 environmental factors and pollutants)?
- 14 ▪ To what extent do receptor surfaces influence the deposition of gases and particles (dry
15 deposition), since dry deposition can contribute significantly to total deposition?
- 16 ▪ To what extent can ecological effects due to NO_x be distinguished from effects due to
17 total reactive nitrogen?
- 18 ▪ Which ecological indicators adequately capture the relationships between ecosystem
19 exposures and responses for the known or anticipated adverse welfare effects that are
20 trying to be protected against?
- 21 ▪ To what extent do the current standards provide protection from the known or anticipated
22 welfare effects associated with NO_x and SO_x?
- 23 ▪ To what extent does the current NO_x standard provide protection against known or
24 anticipated adverse effects associated with total reactive nitrogen?
- 25 ▪ Does the available information provide support for considering different air quality
26 indicators for NO_x and SO_x?
- 27 ▪ Does the available information provide support for the development of appropriate
28 atmospheric deposition transformation functions, and what atmospheric and
29 environmental factors (e.g., co-pollutants, land use) are most appropriate to account for in
30 such a function?

- 1 ▪ Does the available information provide a basis for identifying relevant ecological
2 indicators for the range of ecological endpoints being considered in the review?
- 3 ▪ Does the available information provide support for the development of appropriate
4 ecological effect functions that meaningfully relate to the ecological endpoints being
5 considered, and what ecological factors (e.g., reduced forms of nitrogen, bedrock type,
6 weathering rates) are most relevant for such functions?
- 7 ▪ For which ecological effects being considered is a joint NO_x/SO_x standard most
8 appropriate, and for which ecological effects would separate standards be more
9 appropriate?
- 10 ▪ Is there enough information to determine when ecological effects become adverse?
- 11 ▪ Taking into consideration factors related to determine when the various detrimental
12 ecological effects under consideration occur ; what range of levels, averaging times, and
13 forms of alternative ecological indicators are supported by the information; and what are
14 the uncertainties and limitations in that information?
- 15 ▪ To what extent do specific levels, averaging times, and forms of alternative ecological
16 indicators reduce detrimental impacts attributable to NO_x/SO_x relative to current
17 conditions, and what are the uncertainties in the estimated reductions?

18 In order to answer these questions, the relevant scientific and policy issues that need to be
19 addressed in the science, risk and exposure, and policy assessment portions of this review
20 include the following:

- 21 ▪ Identifying important chemical species in the atmosphere
- 22 ▪ Identifying the atmospheric pathways that govern the chemical transformation, transport,
23 and deposition of total reactive nitrogen and SO_x to the environment
- 24 ▪ Identifying the attributes of ecosystem receptors that govern their susceptibility to effects
25 from deposition of nitrogen and sulfur compounds
- 26 ▪ Identifying the relationships between ambient air quality indicators and ecological
27 indicators of effects (through deposition)
- 28 ▪ Identifying relationships between ecosystem services and ecological indicators

- 1 ▪ Evaluating alternative approaches to assess the adversity of effects on ecosystem
2 services, including, but not limited to, economic valuation
- 3 ▪ Evaluating environmental impacts and sensitivities to varying meteorological scenarios
4 and climate conditions
- 5 ▪ Evaluating the relationship between NO_x and deposition of total reactive nitrogen, and
6 between NO_x and total nitrogen loadings that are related to ecological effects.

7 To the extent the evidence suggests that the current standards do not provide appropriate
8 protection from known or anticipated adverse public welfare effects associated with the criteria
9 pollutants NO_x and SO_x, ecologically meaningful revisions to the current standards will be
10 considered. Recognizing the high degree of complexity that exists in relationships between
11 ambient air concentrations of NO_x and SO_x, deposition of nitrogen and sulfur into sensitive
12 aquatic and terrestrial ecosystems, and associated potential adverse ecological effects, it is
13 anticipated that ecologically meaningful NAAQS need to be structured to take into account such
14 complexity. To provide some context for addressing the key policy-relevant questions that are
15 salient in this review, a possible structure for secondary standards based on meaningful
16 ecological indicators that provides for protection against the range of potentially adverse
17 ecological effects associated with the deposition of NO_x, NH_x, and SO_x has been developed and
18 is shown in **Figure 1.4-1**. In so doing, it was considered how the basic elements of NAAQS
19 standards—indicator, averaging time, form, and level—would be reflected in such a structure.

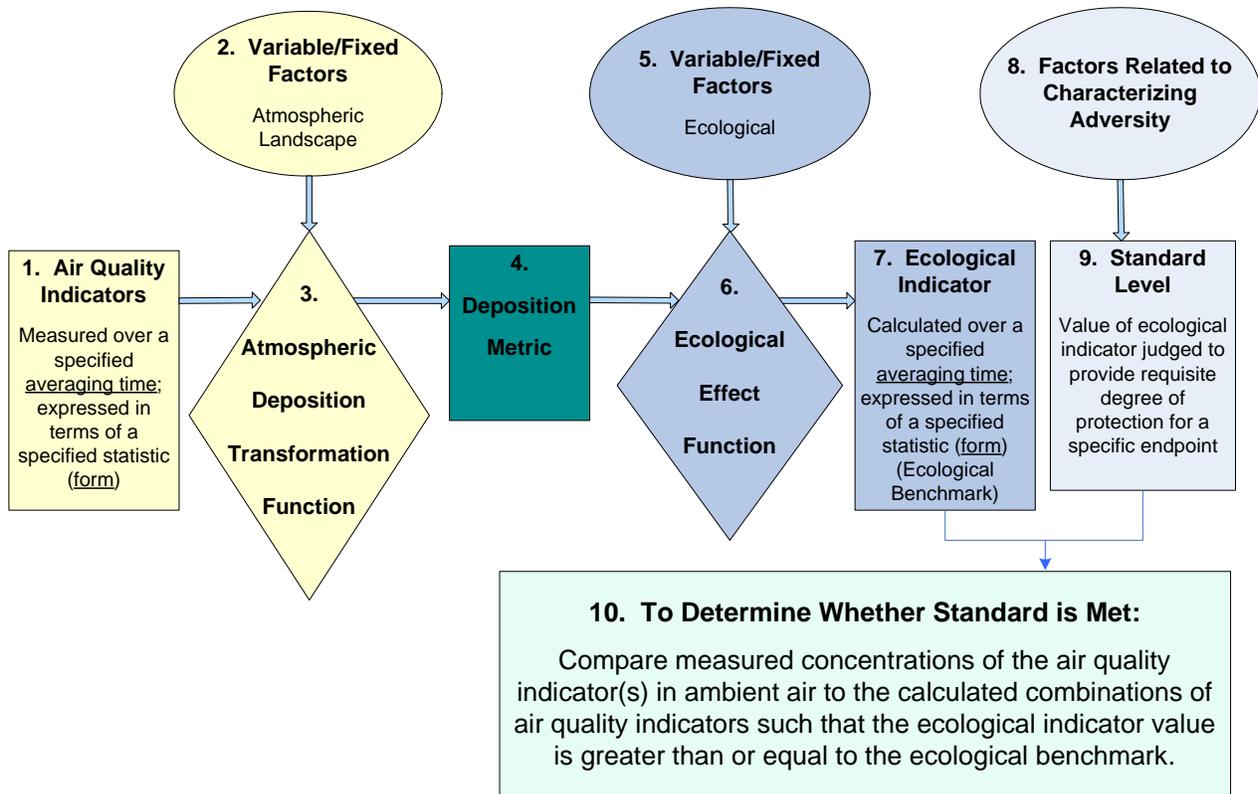


Figure 1.4-1. Possible structure of a secondary NAAQS for NO_x and SO_x based on an ecological indicator.

Figure 1.4-1 illustrates the working structure for an ecological effect-based secondary NAAQS for NO_x and SO_x, together with the combination of various elements that would serve to define such a standard. The subsequent chapters of this report will address each component of this structure. Starting from the left side of **Figure 1.4-1**, Chapter 3 of this report addresses the atmospheric analyses covered in this review, including sources, emissions, concentrations, and deposition and characterization of the spatial and temporal patterns of concentration and deposition in the case study areas (boxes 1 to 4). The Atmospheric Deposition Transformation Function that quantifies the relationship between atmospheric concentrations and deposition of NO_x and SO_x (box 3), while taking atmospheric and landscape factors into account (i.e., deposition velocities, land use, co-pollutants), are addressed in Chapter 3 and Appendix 1 of this report. Chapters 4 and 5 and their associated appendices (Appendices 4-7) focus on the ecological effects of acidification and nutrient enrichment, respectively, and discuss the selection of ecological indicators, ecosystem services, the case study areas and their representativeness, and the evaluation of current conditions in these areas (boxes 4 to 7). For each targeted effect,

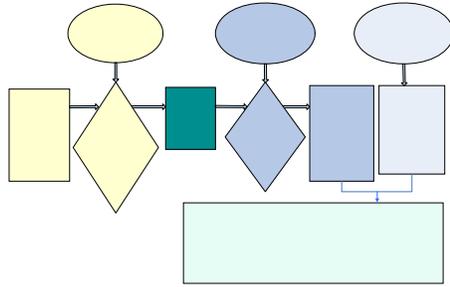
1 the ecological effect functions are derived and described in Chapters 4 and 5 and Appendices 4-7
2 (box 6), and the role of ecosystem services in defining adversity is discussed in Chapters 2, 4 or
3 5, and 7 (box 8). Chapter 7 of this report synthesizes information across different ecological
4 endpoints and identifies impacts linked to ecosystem services that can help to inform the decision
5 as to what levels of ecological indicators are protective against adverse public welfare effects
6 (boxes 7 and 8). All of **Figure 1.4-1** will be evaluated in the policy assessment associated with
7 this review, which will consider the structure of a secondary NAAQS from a statutory standpoint
8 and characterize the atmospheric and ecological inputs discussed throughout the Risk and
9 Exposure Assessment. In addition, the policy assessment will highlight boxes 8, 9, and 10 in
10 **Figure 1.4-1** in a discussion of the risks associated with alternative levels of ecological
11 indicators for each targeted effect area.

12 **1.5 REFERENCES**

- 13 DHEW (U.S. Department of Health, Education, and Welfare). 1970. *Air Quality Criteria for*
14 *Sulfur Oxides*. National Air Pollution Control Administration Publication No. AP-50.
15 Washington, DC: U.S. Government Printing Office.
- 16 Berresheim H; Wine PH; Davis DD. (1995). Sulfur in the atmosphere. In: Singh HB (Ed.),
17 *Composition, chemistry, and climate of the atmosphere* (pp. 251-307). New York, NY:
18 Van Nostrand Reinhold.
- 19 McCurdy, T.R. 1994. *Analysis of High 1-Hour NO₂ Values and Associated Annual Averages*
20 *Using 1988-1992 Data*. Report of the U.S. Environmental Protection Agency, Office of
21 Air Quality Planning and Standards, Durham, NC.
- 22 NAPAP (National Acid Precipitation Assessment Program). 2005. *Report to Congress: An*
23 *Integrated Assessment*. National Acid Precipitation Assessment Program, Washington,
24 DC.
- 25 NAPAP (National Acid Precipitation Assessment Program).1990. *Acidic Deposition: State of*
26 *Science and Technology, Volumes I-IV*. National Acid Precipitation Assessment Program,
27 Washington, DC.

- 1 NRC (National Research Council). 2004. *Air Quality Management in the United States*.
2 Washington, DC: National Academies Press.
- 3 U.S. EPA (Environmental Protection Agency). 1973. *Effects of sulfur oxides in the atmosphere*
4 *on vegetation; revised chapter 5 for air quality criteria for sulfur oxides*. EPA/R3-73-
5 030. U.S. Environmental Protection Agency, Office of Research and Development,
6 Research Triangle Park, NC.
- 7 U.S. EPA (Environmental Protection Agency). 1982. *Air quality criteria for oxides of nitrogen*.
8 EPA-600/8-82-026. U.S. Environmental Protection Agency, Office of Health and
9 Environmental Assessment, Environmental Criteria and Assessment Office, Research
10 Triangle Park, NC.
- 11 U.S. EPA (Environmental Protection Agency). 1984a. *The acidic deposition phenomenon and its*
12 *effects: critical assessment review papers. Volume I: atmospheric sciences*. EPA-600/8-
13 83-016-AF. U.S. Environmental Protection Agency, Office of Research and
14 Development, Washington, DC.
- 15 U.S. EPA (Environmental Protection Agency). 1984b. *The acidic deposition phenomenon and its*
16 *effects: critical assessment review papers. Volume II: effects sciences*. EPA-600/8-83-
17 016-BF. U.S. Environmental Protection Agency, Office of Research and Development,
18 Washington, DC.
- 19 U.S. EPA (Environmental Protection Agency). 1985. *The acidic deposition phenomenon and its*
20 *effects: critical assessment document*. EPA/600/8-85/001. U.S. Environmental Protection
21 Agency, Office of Acid Deposition, Environmental Monitoring, and Quality Assurance,
22 Washington, DC.
- 23 U.S. EPA (Environmental Protection Agency). 1993. *Air quality criteria for oxides of nitrogen*.
24 EPA/600/8-91/049aF-cF. 3v. U.S. Environmental Protection Agency, Office of Health
25 and Environmental Assessment, Environmental Criteria and Assessment Office, Research
26 Triangle Park, NC.

- 1 U.S. EPA (Environmental Protection Agency). 1995a. *Review of the National Ambient Air*
2 *Quality Standards for Nitrogen Dioxide: Assessment of Scientific and Technical*
3 *Information*. OAQPS Staff Paper. EPA-452/R-95-005. U.S. Environmental Protection
4 Agency, Office of Air Quality Planning and Standards, Research Triangle Park, NC.
5 September.
- 6 U.S. EPA (Environmental Protection Agency). 1995b. *Acid Deposition Standard Feasibility*
7 *Study: Report to Congress*. EPA 430-R-95-001a. U.S. Environmental Protection Agency,
8 Office of Air and Radiation, Office of Atmospheric Programs, Acid Rain Division,
9 Washington, DC.
- 10 U.S. EPA (Environmental Protection Agency). 2007. *Integrated Review Plan for the National*
11 *Ambient Air Quality Standards for Particulate Matter*. EPA 452/P-08-006. U.S.
12 Environmental Protection Agency, National Center for Environmental Assessment,
13 Office of Research and Development, and Health and Environmental Impacts Division,
14 Office of Air Quality Planning and Standards, Office of Air and Radiation, Research
15 Triangle Park, NC. October.
- 16 U.S. EPA (Environmental Protection Agency). 2008. *Integrated Science Assessment (ISA) for*
17 *Oxides of Nitrogen and Sulfur—Ecological Criteria (Final Report)*. EPA/600/R-
18 08/082F. U.S. Environmental Protection Agency, National Center for Environmental
19 Assessment—RTP Division, Office of Research and Development, Research Triangle
20 Park, NC. Available at <http://cfpub.epa.gov/ncea/cfm/recordisplay.cfm?deid=201485>.
- 21 Wolff, G.T. 1993. On a NO_x-focused control strategy to reduce O₃. *Journal of the Air and Waste*
22 *Management Association* 43:1593–1596.
- 23 Wolff, G.T. 1995. CASAC closure letter for the 1995 OAQPS Staff Paper. Letter sent to Carol
24 M. Browner, Administrator, U.S. Environmental Protection Agency. August 22.
- 25

1
2

3 2.0 OVERVIEW OF RISK AND EXPOSURE ASSESSMENT

4 2.1 INTRODUCTION

5 The Risk and Exposure Assessment focuses on ecosystem welfare effects that result from
6 the deposition of total reactive nitrogen and sulfur. Because ecosystems are diverse in biota,
7 climate, geochemistry, and hydrology, response to pollutant exposures can vary greatly between
8 ecosystems. In addition, these diverse ecosystems are not distributed evenly across the United
9 States. To target nitrogen and sulfur acidification and nitrogen and sulfur enrichment, the Risk
10 and Exposure Assessment addresses four main targeted ecosystem effects on terrestrial and
11 aquatic systems identified by the *Integrated Science Assessment (ISA) for Oxides of Nitrogen
12 and Sulfur–Ecological Criteria (Final Report)* (ISA; U.S. EPA, 2008a):

- 13 ■ Aquatic acidification due to nitrogen and sulfur
- 14 ■ Terrestrial acidification due to nitrogen and sulfur
- 15 ■ Aquatic nutrient enrichment, including eutrophication
- 16 ■ Terrestrial nutrient enrichment.

17 In addition to these four targeted ecosystem effects, this assessment qualitatively
18 addresses the influence of sulfur oxides (SO_x) deposition on methylmercury production, nitrous
19 oxide (N₂O) effects on climate, and nitrogen effects on primary productivity and biogenic
20 greenhouse gas fluxes, and phytotoxic effects on plants.

21 Because the targeted ecosystem effects outlined above are not evenly distributed across
22 the United States, the Risk and Exposure Assessment identified case studies for the analyses
23 based on ecosystems identified as sensitive to nitrogen and/or sulfur deposition effects. This Risk
24 and Exposure Assessment builds upon the scientific information presented in the ISA, with

1 ecological indicator(s) and case study areas selected based on the information presented (U.S.
2 EPA, 2008a).

3 This assessment builds upon the scientific information presented in the ISA (U.S. EPA,
4 2008a) from which ecological indicator(s) and case study areas were selected. Eight case study
5 areas and two supplemental study areas (Rocky Mountain National Park and Little Rock Lake,
6 WI) are summarized in **Table 2.1-1** based on ecosystem characteristics, indicators, and
7 ecosystem service information developed for this Risk and Exposure Assessment. Detailed
8 explanations of this information are presented in Chapters 4 and 5 of this report (i.e., *Risk and*
9 *Exposure Assessment for Review of the Secondary National Ambient Air Quality Standards for*
10 *Oxides of Nitrogen and Oxides of Sulfur*), and a map highlighting each of 8 the case study areas
11 and the Rocky Mountain National Park is shown in **Figure 2.1-1**.

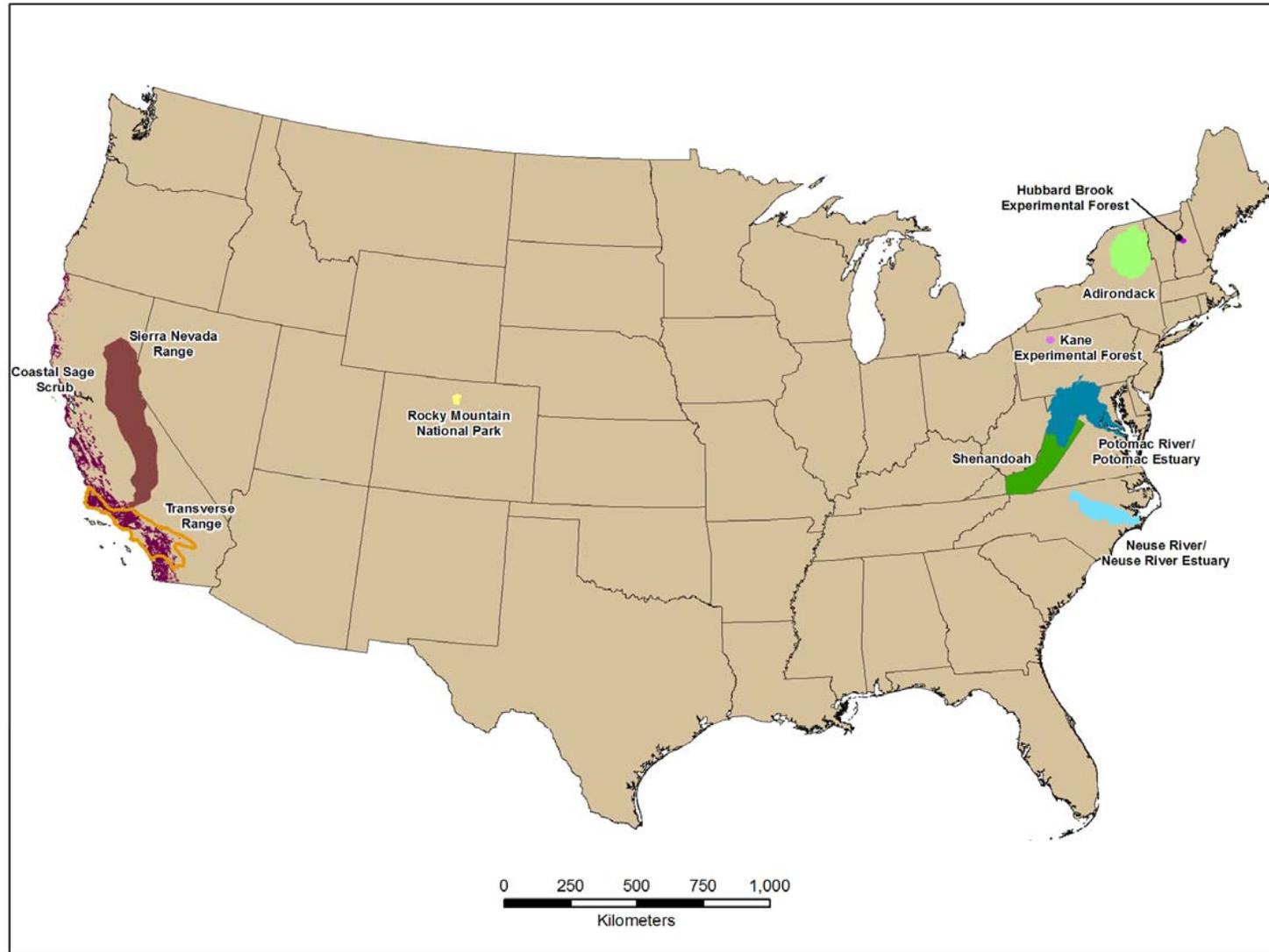
1 **Table 2.1-1.** Summary of Sensitive Characteristics, Indicators, Effects, and Impacted Ecosystem Services Analyzed for Each Case
 2 Study Evaluated in This Review

Targeted Ecosystem Effect	Characteristics of Sensitivity (Variable Ecological Factors)	Biological/ Chemical Indicator	Ecological Endpoint	Ecological Effects	Ecosystem Services Impacted	Case Study Areas
Aquatic Acidification	Geology, surface water flow, soil depth, weathering rates	Al pH ANC	Species richness, abundance, composition, ANC	Species losses of fish, phytoplankton, and zooplankton; changed community composition, ecosystem structure, and function	Subsistence fishing, recreational fishing, other recreational activities	Adirondack Mountains, NY (referred to as Adirondack) Shenandoah National Park, VA (referred to as Shenandoah)
Terrestrial Acidification	Geology, surface water flow, soil depth, weathering rates	Soil base saturation Al Ca C:N ratio	Tree health of red spruce and sugar maple, ANC, base cation :Al ratio	Decreased tree growth, increased susceptibility to stress, episodic dieback; changed community composition, ecosystem structure, and function	Provision of food and wood products, recreational activities, natural habitat, soil stabilization, erosion control, water regulation, climate regulation	Kane Experimental Forest (Allegheny Plateau, PA) Hubbard Brook Experimental Forest (White Mountains, NH)
Aquatic Nutrient Enrichment	nitrogen-limited systems, presence of nitrogen in surface water, eutrophication status, nutrient criteria	Chlorophyll <i>a</i> , macroalgae, dissolved oxygen, nuisance/toxic algal blooms, submerged aquatic vegetation (SAV)	Changes in Eutrophication Index (EI)	Habitat degradation, algal blooms, toxicity, hypoxia, anoxia, fish kills, decreases in biodiversity	Commercial and recreational fishing, other recreational activities, aesthetic value, nonuse value flood and erosion control	Potomac River Basin, Chesapeake Bay (referred to as Potomac River/Potomac Estuary) Neuse River Basin, Pamlico Sound (referred to as Neuse River/Neuse River Estuary)

Targeted Ecosystem Effect	Characteristics of Sensitivity (Variable Ecological Factors)	Biological/ Chemical Indicator	Ecological Endpoint	Ecological Effects	Ecosystem Services Impacted	Case Study Areas
Terrestrial Nutrient Enrichment	Presence of acidophytic lichens, anthropogenic land cover	Cation exchange capacity, C:N ratios, Ca:Al ratios, NO ₃ ⁻ leaching and export	Species composition, lichen presence/absence, soil root mass changes, NO ₃ breakthrough to water, biomass	Species changes, nutrient enrichment of soil, changes in fire regime, changes in nutrient cycling	Recreation, aesthetic value, nonuse value, fire regulation, loss of habitat, loss of biodiversity, water quality	Coastal Sage Scrub (southern, coastal California) and Mixed Conifer Forest (San Bernardino Mountains of the Transverse Range and Sierra Nevada Mountain Ranges, California); Rocky Mountain National Park (a supplemental study area)

1 **Note:** ANC = acid neutralizing capacity, SAV = submerged aquatic vegetation, EI = eutrophication index.

2



3 **Figure 2.1-1.** National map highlighting the 8 case study areas and the Rocky Mountain National Park (a supplemental
4 study area) evaluated in the Risk and Exposure Assessment.

1 To address the policy-relevant questions that guide the scope of this review, the Risk and
2 Exposure Assessment evaluates the relationships between atmospheric concentrations,
3 deposition, biologically relevant exposures, targeted ecosystem effects, and ecosystem services.
4 To evaluate the nature and magnitude of ecosystem responses associated with adverse effects,
5 the Risk and Exposure Assessment examines various ways to quantify the relationships between
6 air quality indicators, deposition of biologically available forms of nitrogen and sulfur,
7 ecologically relevant indicators relating to deposition, exposure and effects on sensitive
8 receptors, and related effects resulting in changes in ecosystem structure and services. The intent
9 of the Risk and Exposure Assessment is to determine the exposure metrics that incorporate the
10 temporal considerations (i.e., biologically relevant timescales), pathways, and ecologically
11 relevant indicators necessary to maintain the functioning of these ecosystems. To the extent
12 feasible, this assessment evaluates the overall load to the system for nitrogen and sulfur, as well
13 as the variability in ecosystem responses to these pollutants. In addition, the assessment
14 evaluates the contributions of atmospherically deposited nitrogen and sulfur relative to total
15 loadings in the environment. Since oxidized nitrogen is the listed criteria pollutant (currently
16 measured by the ambient air quality indicator NO₂) for the atmospheric contribution to total
17 nitrogen, this assessment examines the contribution of nitrogen oxides (NO_x) to total reactive
18 nitrogen in the atmosphere, relative to the contributions of reduced forms of nitrogen (e.g.,
19 ammonia, ammonium), to ultimately assess how a meaningful secondary National Ambient Air
20 Quality Standards (NAAQS) might be structured.

21 The Risk and Exposure Assessment for the secondary NAAQS review for NO_x and SO_x
22 will aid the Administrator in judging whether the current secondary standards are requisite to
23 protect public welfare from any known or anticipated adverse effects, or whether these standards
24 should be retained, revised, revoked, and/or replaced with alternative standard(s) to provide the
25 required protection.

26 Previous reviews of secondary NAAQS have characterized adversity according to the
27 ecological effects associated with that pollutant. For example, in the previous ozone (O₃)
28 secondary NAAQS review, biomass loss and foliar injury were the main effects determining
29 adversity to public welfare on public lands, while in the particulate matter (PM) secondary
30 NAAQS review, the loss of visibility was used. There is an important distinction between a
31 scientifically defined and documented adverse effect to a given ecological system or ecological

1 endpoint and an adverse impact on public welfare from a statutory perspective. While adverse
2 effects to ecosystems from a scientific perspective will be used to inform the Administrator’s
3 decision, the degree of change in an ecological indicator or service that corresponds to an
4 adverse public welfare effect is ultimately decided by the Administrator.

5 For assessing this set of secondary NAAQS, in addition to assessing the degree of
6 scientific impairment of ecological systems relating to inputs of NO_x and sulfur oxides (SO_x),
7 this Risk and Environmental Assessment presents a broad look into the concept of ecosystem
8 services that is one tool that can help link what is considered to be a biologically adverse effect
9 with a known or anticipated adverse effect to public welfare through ecosystem services.

10 In this Risk and Exposure Assessment, ecosystem services is used as an umbrella term to
11 aid in describing the impacts of ecological effects on public welfare and to help explain how
12 these effects are viewed by the public. Ecosystem services are addressed in more detail in
13 Section 2.4 of this chapter, throughout the case study analyses in Chapters 4 and 5, and in the
14 examination of the structure of an ecologically meaningful secondary standard in the policy
15 assessment document. The ability to inform decisions on the level of a secondary NAAQS will
16 require the development of clear linkages between biologically adverse effects and effects that
17 are adverse to public welfare as related to ecosystem services. The concept of adversity to public
18 welfare does not require the use of ecosystem services, yet it is envisioned as a beneficial tool for
19 this review that may provide more information on the linkages between adverse ecological
20 effects and adverse public welfare effects.

21 **2.2 SEVEN-STEP APPROACH**

22 The seven basic steps guiding the overall Risk and Exposure Assessment and the
23 assessments for each case study area of interest are highlighted below. These steps were initially
24 presented in the scope and methods plan for this review (U.S. EPA, 2008b) and received Clean
25 Air Scientific Advisory Committee (CASAC) approval; therefore, this approach is being carried
26 forward for the Risk and Exposure Assessment. The seven steps address the selection of the
27 targeted ecosystem effects, indicators, and ecosystem services measured for exposure via
28 atmospheric deposition of total reactive nitrogen and sulfur from ambient air. The initial step of
29 identifying effects, sensitive ecosystems, and potential indicators is documented in the ISA (see
30 Chapter 3, U.S. EPA, 2008a). In addition, the ISA identifies and reviews candidate multimedia

1 models available for fate and transport analyses of a variety of ecosystems. The science
2 documented in the ISA provides critical inputs into the Risk and Exposure Assessment. For some
3 of the desired case study areas, data were not abundant enough to perform a quantitative
4 assessment for each of the steps; in those cases, some steps have been executed in a qualitative or
5 semiquantitative fashion.

6 The details of the seven steps are addressed in each case study description. The steps are
7 as follows:

- 8 ▪ **Step 1.** Plan for assessment using documented effects, such as biological, chemical, and
9 ecological indicators; ecological responses; and potential ecosystem services.
- 10 ▪ **Step 2.** Map characteristics of sensitive areas that show ecological responses using
11 research findings and geographic information systems (GIS) mapping.
- 12 ▪ **Step 3.** Select risk and exposure case study assessment area(s) within a sensitive area.
- 13 ▪ **Step 4.** Evaluate current loads and effects to case study assessment areas, including
14 ecosystem services, where possible.
- 15 ▪ **Step 5.** Evaluate representativeness of case study areas to larger sensitive areas.
- 16 ▪ **Step 6.** Assess the current ecological conditions for those larger sensitive areas.
- 17 ▪ **Step 7.** Develop ecological effect functions for the targeted ecosystem effects (e.g.,
18 aquatic acidification).

19 The policy assessment for this review addresses the characterization of risks associated
20 with alternative levels of ecological indicators and the associated impacts on ecosystem services.

2.3 LINKAGES FOR STRUCTURING ECOLOGICALLY RELEVANT STANDARDS

The framework shown in **Figure 2.3-1** depicts an example of how an ecologically meaningful secondary NAAQS might be structured. This example presents a system of linked functions that translate an air quality indicator (e.g., concentrations of NO_x and SO_x) into an ecological indicator that expresses either the potential for deposition of nitrogen and sulfur to acidify an ecosystem or for nitrogen to adversely enrich an ecosystem. This system encompasses the linkages between ambient air concentrations and resulting deposition metrics, as well as between the deposition metric and the ecological indicator of concern. For example, the atmospheric deposition transformation function (box 3) translates ambient air concentrations of NO_x and SO_x to nitrogen and sulfur deposition metrics, while the ecological effect function (box 6) relates the deposition metric into the ecological indicator.

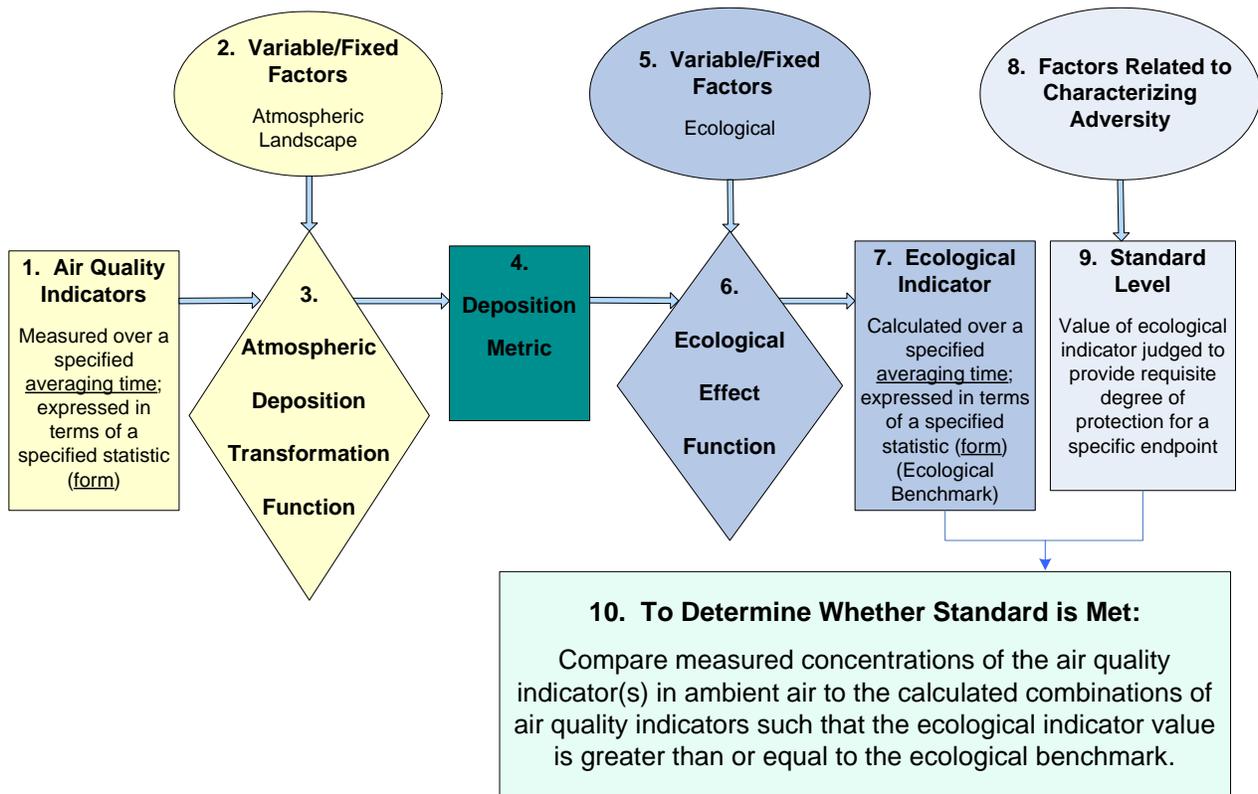


Figure 2.3-1. Possible structure of a secondary NAAQS for NO_x and SO_x based on an ecological indicator.

1 The amounts of NO_x and SO_x in the ambient air can be used to derive a deposition metric
2 (via the atmospheric deposition transformation function), which can then be used to derive a
3 level of an ecological indicator (through the ecological effect function) that falls within the range
4 defined as acceptable by the standard; by definition, the levels of NO_x and SO_x will be
5 considered to meet that standard of protection. The atmospheric levels of NO_x and SO_x that
6 satisfy a particular level of ecosystem protection are those levels that result in an amount of
7 deposition that is less than the amount of deposition a given ecosystem can accept without
8 degradation of the ecological indicator for a targeted ecosystem effect. This latter amount is
9 referred to as the maximum depositional load and is the amount that solves a mass-balance
10 equation for a given ecological indicator.

11 Modifying factors that alter the relationship between ambient air concentrations of NO_x
12 and SO_x and depositional loads of nitrogen and sulfur, and those that modify the relationship
13 between depositional loads and the ecological indicator, are discussed more fully throughout the
14 discussion of atmospheric analyses in Chapter 3 of this report, the review of case study analyses
15 in Chapters 4 and 5, and the overview of the synthesis to inform the standard-setting process
16 provided in Chapter 7. In addition, the role of ecosystem services in determining an adverse
17 effect to public welfare is introduced in Section 2.4 and highlighted throughout the case study
18 analyses in Chapters 4 and 5. This role will be discussed in the policy assessment document
19 when characterizing risks associated with the development of a standard(s).

20 **2.4 ECOSYSTEM SERVICES**

21 The Risk and Exposure Assessment evaluates the benefits received from the resources
22 and processes that are supplied by ecosystems. Collectively, these benefits are known as
23 ecosystem services and include products or provisions, such as food and fiber; processes that
24 regulate ecosystems, such as carbon sequestration; cultural enrichment; and supportive processes
25 for services, such as nutrient cycling. Ecosystem services are distinct from other ecosystem
26 products and functions because there is human demand for these services.

27 In the Millennium Ecosystem Assessment (MEA), ecosystem services are classified into
28 four main categories:

- 29 ▪ **Provisioning.** Includes products obtained from ecosystems, such as the production of
30 food and water.

- 1 ▪ **Regulating.** Includes benefits obtained from the regulation of ecosystem processes, such
- 2 as the control of climate and disease.
- 3 ▪ **Cultural.** Includes the nonmaterial benefits that people obtain from ecosystems through
- 4 spiritual enrichment, cognitive development, reflection, recreation, and aesthetic
- 5 experiences.
- 6 ▪ **Supporting.** Includes those services necessary for the production of all other ecosystem
- 7 services, such as nutrient cycles and crop pollination (MEA, 2005a).

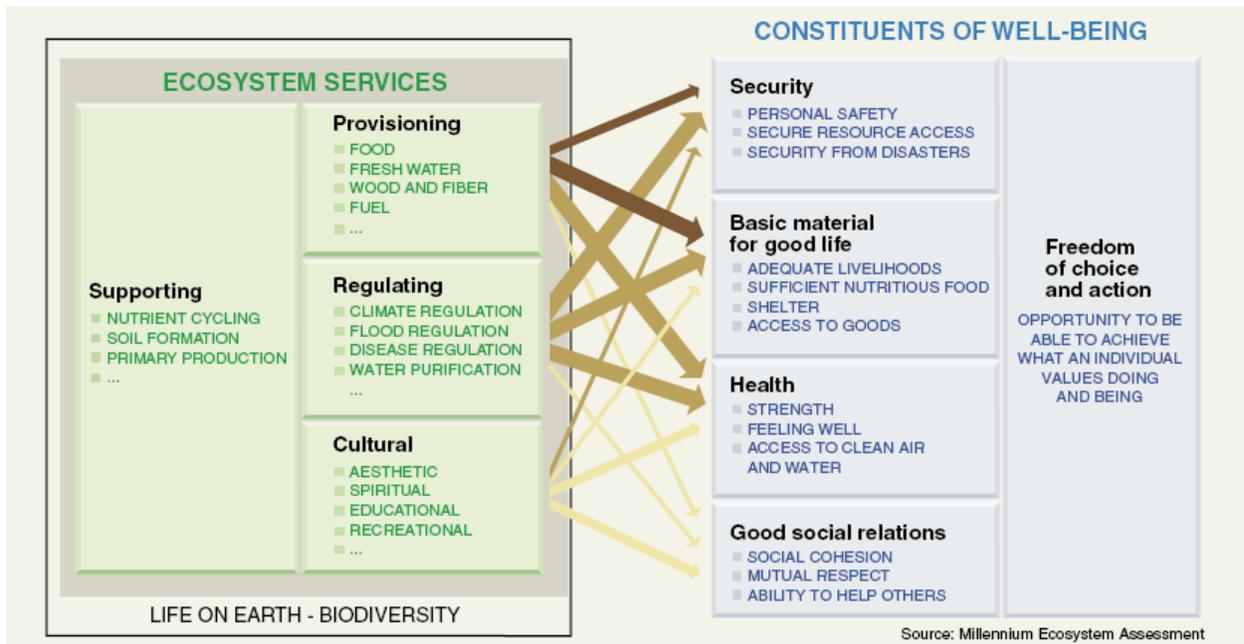
8 **Figure 2.4-1** provides the World Resources Institute’s schematic demonstrating the

9 connections between the categories of ecosystem services and human well-being. The

10 interrelatedness of these categories means that any one ecosystem may provide multiple services.

11 Changes in these services can impact human well-being by affecting security, health, social

12 relationships, and access to basic material goods (MEA, 2005b).



13

14
15

Figure 2.4-1. Millennium ecosystem assessment categorization of ecosystem services and their links to human well-being (MEA, 2005a).

16
17
18
19

Historically, ecosystem services have been undervalued and overlooked; however, more recently, the degradation and destruction of ecosystems has piqued interest in assessing the value of these services. In addition, valuation may be an important step from a policy perspective because it can be used to compare the costs and benefits of altering versus maintaining an

1 ecosystem (i.e., it may be easier to protect than repair ecosystem effects). In this Risk and
2 Exposure Assessment, valuation is used, where possible, based on available data in the case
3 study areas.

4 The economic approach to the valuation of ecosystem services is laid out as follows in
5 EPA's *Ecological Benefits Assessment Strategic Plan*: "Economists generally attempt to estimate
6 the value of ecological goods and services based on what people are willing to pay (WTP) to
7 increase ecological services or by what people are willing to accept (WTA) in compensation for
8 reductions in them" (U.S. EPA, 2006). There are three primary approaches for estimating the
9 value of ecosystem services: market-based approaches, revealed preference methods, and stated
10 preference methods (U.S. EPA, 2006). Because economic valuation of ecosystem services can be
11 difficult, nonmonetary valuation using biophysical measurements and concepts also can be used.
12 Examples of nonmonetary valuation methods include the use of relative-value indicators (e.g., a
13 flow chart indicating uses of a waterbody, such as boatable, fishable, swimmable); another
14 assigns values to ecosystem goods and services through the use of the common currency of
15 energy. Valuation may be an important step from a policy perspective because it can be used to
16 compare the costs and benefits of altering versus maintaining an ecosystem (i.e., it may be easier
17 to protect than repair ecosystem effects). In this review, valuation is used, where possible, based
18 on available data in the case study areas.

19 The ecosystems of interest in this Risk and Exposure Assessment are heavily impacted by
20 the effects of anthropogenic air pollution, which may alter the services provided by the
21 ecosystems in question. For example, changes in forest health as a result of soil acidification
22 from NO_x and SO_x deposition may affect supporting services such as nutrient cycling;
23 provisioning services such as timber production; and regulating services such as climate
24 regulation. In addition, eutrophication caused by NO_x deposition may affect supporting services
25 such as primary production; provisioning services such as food; and cultural services such as
26 recreation and ecotourism.

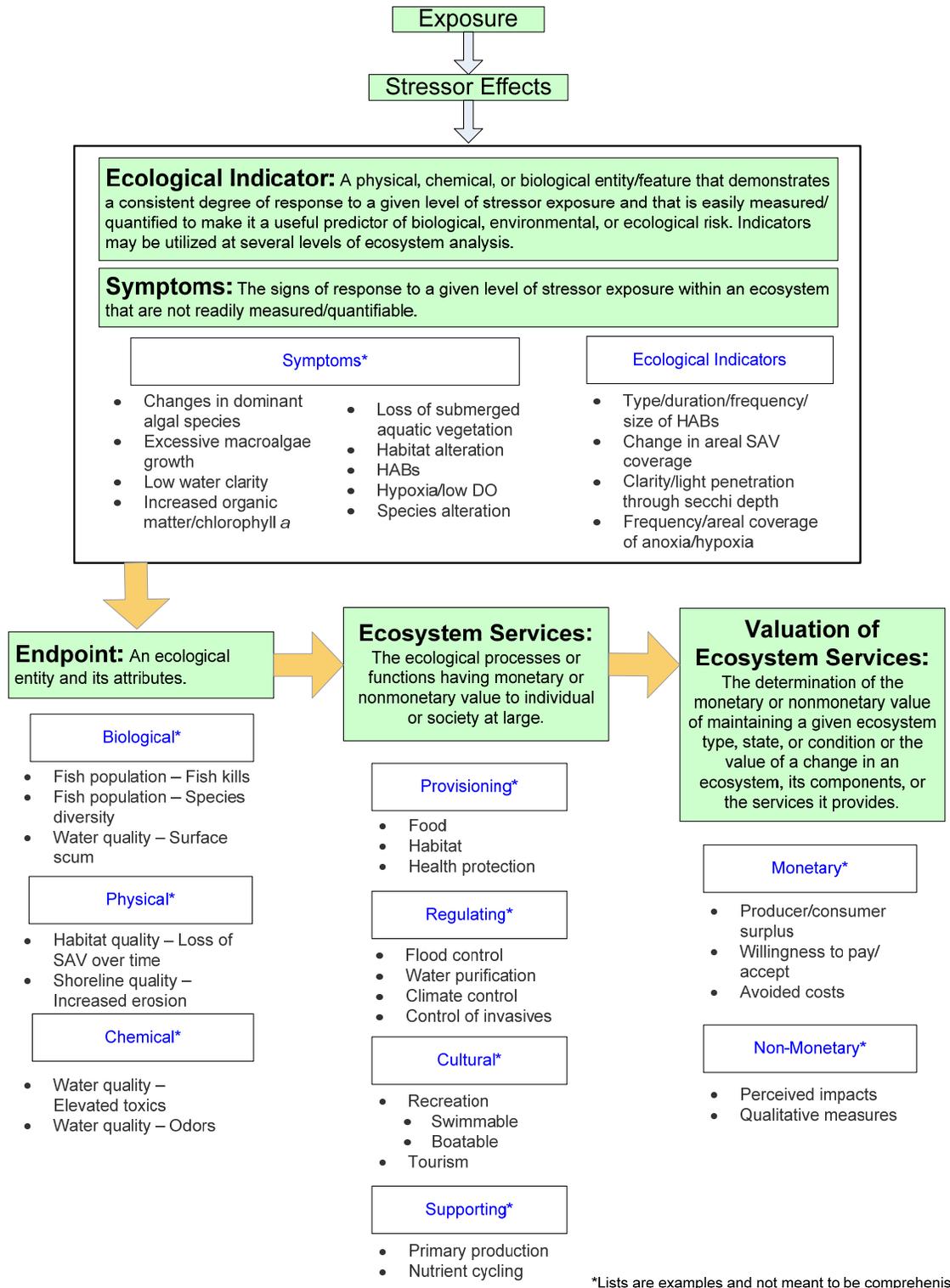
27 Where possible, linkages to ecosystem services from indicators of each effect identified
28 in Step 1 of the Risk and Exposure Assessment were developed. These linkages were based on
29 existing literature and models, focus on the services identified in the peer-reviewed literature,
30 and are essential to any attempt to evaluate air pollution-induced changes in the quantity and/or
31 quality of ecosystem services provided. According to EPA's Science Advisory Board Committee

1 on Valuing the Protection of Ecological Systems and Services, these linkages are critical
2 elements for determining the valuation of benefits of EPA-regulated air pollutants (SAB C-
3 VPESS, 2007). **Figure 2.4-2** provides an example pathway for nitrogen deposition in an aquatic
4 ecosystem that links the ecological endpoints to changes in services and, finally, to valuation.

5 This Risk and Exposure Assessment identifies the primary ecosystem service(s) for both
6 acidification and enrichment and for the targeted ecosystem effects under consideration in this
7 exposure assessment (see **Table 2.1-1**). Examples of some of the linkages between impacts and
8 each targeted ecosystem effect in relation to specific ecosystem services are summarized below
9 and in **Table 2.4-1**.

10

Aquatic Enrichment Example



1
2 **Figure 2.4-2.** Pathway from nitrogen deposition to valuation for an aquatic system.
3 **Note:** HABs = harmful algal blooms, DO = dissolved oxygen , SAV = submerged
4 aquatic vegetation.

1 **2.4.1 Aquatic Acidification**

2 The analysis of ecosystem services for the aquatic acidification focused on recreational
3 fishing. Fish abundance (decreased species richness) has been quantitatively linked to
4 acidification through monitoring data and modeling of acid neutralizing capacity. Relevant
5 ecosystem services were quantified, and values were estimated using a Random Utility model for
6 fishing services and contingent valuation studies to estimate gains in total services provided by
7 the Adirondack and New York State lakes case study area.

8 **2.4.2 Terrestrial Acidification**

9 The ecosystem services analysis for Terrestrial Acidification Case Study concentrated on
10 the provision of food and wood products and on recreational activity. Sugar maple and red
11 spruce abundance and growth (i.e., crown vigor, biomass, and geographic extent) were
12 quantitatively linked to acidification symptoms through the Forest Inventory and Analysis
13 National Program (FIA) database analyses. Results of the FIA database analysis were input to
14 the Forest and Agriculture Sector Optimization Model – Green House Gas version
15 (FASOMGHG) to estimate producer and consumer surplus gains associated with decreased
16 acidification.

17 **2.4.3 Aquatic Nutrient Enrichment**

18 The ecosystem services analysis for aquatic nutrient enrichment evaluated several
19 cultural ecosystem services, including recreational fishing, boating, and beach use. In addition,
20 aesthetic and nonuse values were evaluated; the impacts on recreational fishing (e.g., closings,
21 decreased species richness) to eutrophication symptoms through monitoring data were
22 quantitatively linked; other recreational activities and aesthetic and non-use services to
23 eutrophication symptoms were quantitatively related through user surveys and valuation
24 literature; and the current commercial fishing markets were described. Although little data is
25 available to link any decrease in commercial landings or subsistence fishing directly to
26 eutrophication, it seems likely that these activities would be impacted.

27 **2.4.4 Terrestrial Nutrient Enrichment**

28 The ecosystem services analysis for terrestrial nutrient enrichment for the coastal sage
29 scrub and mixed conifer forest ecosystems focused on services such as recreation, aesthetic, and

1 non-use services, including existence values. Given the lack of data available to develop a
2 quantitative analysis of service impacts, the impacts on these ecosystems are addressed in a
3 qualitative fashion.

4 **2.4.5 Sulfur and Mercury Methylation**

5 The major ecosystem services potentially impacted by mercury methylation are
6 provisioning and cultural services. Fishing and shellfishing can involve both commercial
7 operations and sport fishing, both of which provide food for human populations. For some socio-
8 economic groups (especially low-income groups), fishing is a subsistence activity that makes a
9 significant contribution to household food intake. Sport fishing often involves important
10 recreational services, and for many groups (e.g., Native Americans, Alaska Native villagers),
11 fishing and consuming local fish or shellfish is of cultural and spiritual significance. A synthesis
12 of the ecosystem service and valuation aspects of fishing and shellfishing activities, with a focus
13 on the mercury pollution issues affecting human health and well-being, is found in the
14 *Regulatory Impact Analysis of the Clean Air Mercury Rule* (U.S. EPA, 2005) and in the *Mercury*
15 *Study Report to Congress* (U.S. EPA, 1997).

16

1 **Table 2.4-1.** Ecological Impacts Associated with Acidification, Nutrient Enrichment, and Increased Mercury Methylation and Their
 2 Associated Ecosystem Services

Targeted Ecosystem Effect	Provisioning Services	Regulating Services	Cultural Services	Supporting Services
Aquatic Acidification	<ul style="list-style-type: none"> ▪ Fishing (subsistence) 	<ul style="list-style-type: none"> ▪ Biological control 	<ul style="list-style-type: none"> ▪ Recreational fishing ▪ Nonuse 	Not Available
Terrestrial Acidification	<ul style="list-style-type: none"> ▪ Food, wood products 	<ul style="list-style-type: none"> ▪ Erosion control ▪ Fire regulation ▪ Hydrologic ▪ Climate 	<ul style="list-style-type: none"> ▪ Recreational activity ▪ Aesthetic ▪ Nonuse 	Not Available
Aquatic Nutrient Enrichment	<ul style="list-style-type: none"> ▪ Commercial fishing 	<ul style="list-style-type: none"> ▪ Erosion control ▪ Flood control 	<ul style="list-style-type: none"> ▪ Recreational activity ▪ Aesthetic ▪ Nonuse 	<ul style="list-style-type: none"> ▪ Nutrient cycling
Terrestrial Nutrient Enrichment				
Coastal Sage Scrub	Not Available	<ul style="list-style-type: none"> ▪ Fire regulation ▪ Hydrologic control ▪ Climate 	<ul style="list-style-type: none"> ▪ Recreational activity ▪ Aesthetic ▪ Nonuse 	Not Available
Mixed Conifer Forest	Not Available	<ul style="list-style-type: none"> ▪ Hydrologic control ▪ Climate 	<ul style="list-style-type: none"> ▪ Recreational activity ▪ Aesthetic ▪ Nonuse 	<ul style="list-style-type: none"> ▪ Nutrient cycling,
Sulfur and Mercury Methylation	<ul style="list-style-type: none"> ▪ Commercial and subsistence fishing 	Not Available	<ul style="list-style-type: none"> ▪ Recreational fishing ▪ Nonuse 	Not Available

1 **2.5 UNCERTAINTY**

2 The scope of this Risk and Exposure Assessment involves quantifying a number of
3 relationships along the path of moving from ambient concentrations of NO_x, NH_x, and SO_x to
4 their transformation products and deposition in the environment. The environmental effects of
5 nitrogen and sulfur deposition vary widely and the extent of these effects in time and space is
6 often uncertain in both terrestrial and aquatic ecosystems. The relationships between deposition,
7 ecological effects, ecological indicators, and ecosystem services are also quantified. Uncertainty
8 and variability are present at each step in this framework (as shown in **Figure 2.3-1**). In addition,
9 extrapolating from a case study area to a larger assessment area introduces additional uncertainty
10 and potential error into the process. Understanding the nature, sources, and importance of these
11 uncertainties will help inform the standard setting process in the policy assessment phase of this
12 review.

13 Uncertainty represents a lack of knowledge about the true value of a parameter that can
14 result from inadequate or imperfect measurement. Uncertainty can be reduced by obtaining
15 additional measurements, data, and information. Conceptual and numerical uncertainty can be
16 bounded by testing a range of inputs and parameters in atmospheric and ecological numerical
17 process models, like the ones used in this assessment. An additional source of uncertainty is error
18 due to the use of incorrect measurements, methods, data, or models. Error can be identified and
19 addressed by thorough evaluation, review, and consultation with outside experts.

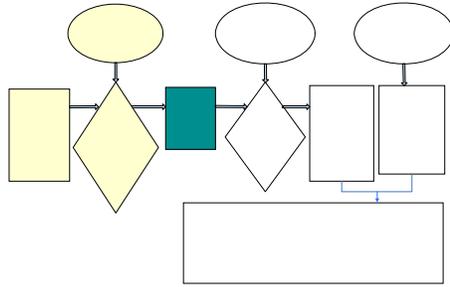
20 Variability in space and time is a component of all environmental systems and represents
21 actual differences in the value of a parameter or attribute of an ecological indicator. Variability
22 describes the natural variation in a system and cannot be reduced by taking additional
23 measurements of a parameter, although it is possible to characterize the range of variation in a
24 measurement or parameter. For example, there is natural variability among similar ecosystems
25 nationwide, some of which are more sensitive to acidification and/or nutrient enrichment than
26 others, just as there is natural variability in the precipitation amounts that produce wet deposition
27 loadings to these systems.

28 Selected terms and sources of uncertainty and variability are discussed, as appropriate, in
29 each section of this Risk and Exposure Assessment.

1 **2.6 REFERENCES**

- 2 MEA (Millennium Ecosystem Assessment Board). 2005a. *Ecosystems and Human Well-being:*
3 *Current State and Trends, Volume 1.* Edited by R. Hassan, R. Scholes, and N. Ash.
4 Washington: Island Press. Available at [http://www.millenniumassessment.org/](http://www.millenniumassessment.org/documents/document.766.aspx.pdf)
5 [documents/document.766.aspx.pdf](http://www.millenniumassessment.org/documents/document.766.aspx.pdf).
- 6 MEA (Millennium Ecosystem Assessment Board). 2005b. *Millennium Ecosystem Assessment*
7 *Reports.* Washington: Island Press. Available at
8 <http://www.millenniumassessment.org/en/index.aspx>.
- 9 SAB C-VPES (Science Advisory Board Committee on Valuing the Protection of Ecological
10 Systems and Services). 2007. *Valuing the Protection of Ecological Systems and Services.*
11 SAB CVPES Draft Report, September 24.
- 12 U.S. EPA (Environmental Protection Agency) 1997. *Mercury Study Report to Congress.* EPA-
13 452/R-97-003. Office of Air Quality Planning & Standards and Office of Research and
14 Development, Washington, DC. December.
- 15 U.S. EPA (Environmental Protection Agency) 2005. *Regulatory Impact Analysis of the Final*
16 *Clean Air Mercury Rule.* U.S. Environmental Protection Agency, Office of Air Quality
17 Planning and Standards Air Quality Strategies and Standards Division. Research Triangle
18 Park, NC. EPA-452/R-05-003. March 2005
- 19 U.S. EPA (Environmental Protection Agency). 2006. *Ecological Benefits Assessment Strategic*
20 *Plan.* EPA-240-R-06-001. Office of the Administrator. Washington, DC. Available at
21 <http://www.epa.gov/economics>.
- 22 U.S. EPA (Environmental Protection Agency). 2008a. *Integrated Science Assessment (ISA) for*
23 *Oxides of Nitrogen and Sulfur–Ecological Criteria (Final Report).* EPA/600/R-
24 08/082F. U.S. Environmental Protection Agency, National Center for Environmental
25 Assessment–RTP Division, Office of Research and Development, Research Triangle
26 Park, NC. Available at <http://cfpub.epa.gov/ncea/cfm/recordisplay.cfm?deid=201485>.

- 1 U.S. EPA (Environmental Protection Agency). 2008b. Draft Scope and Methods Plan for
- 2 Risk/Exposure Assessment: Secondary NAAQS Review for Oxides of Nitrogen and
- 3 Oxides of Sulfur. U.S. Environmental Protection Agency, Office of Air Quality Planning
- 4 and Standards, Research Triangle Park, NC. EPA-452/D-08-002. March 2008.
- 5



1
2
3
4
5
6
7
8
9
10
11
12
13
14
15
16
17
18
19
20
21
22

3.0 SOURCES, AMBIENT CONCENTRATIONS, AND DEPOSITION

This chapter discusses current emissions sources of nitrogen and sulfur, as well as atmospheric concentrations, estimates of deposition, policy-relevant background, and non-ambient loadings of nitrogen and sulfur to ecosystems. Both measured and modeled data are used to evaluate current contributions of nitrogen and sulfur compounds to the Risk and Exposure Assessment case study areas. The case study areas are 1) Adirondack Mountains (referred to as Adirondack); 2) Blue Ridge Mountains/Shenandoah National Park, Virginia (referred to as Shenandoah); 3) Kane Experimental Forest (KEF) on the Allegheny Plateau of Pennsylvania; 4) Hubbard Brook Experimental Forest (HBEF) in the White Mountains of New Hampshire; 5) Potomac River/Potomac Estuary; 6) Neuse River/Neuse River Estuary; 7) southern California Coastal Sage Scrub (CSS); and 8) Pacific coast states’ Mixed Conifer Forest (MCF), including the Transverse (or Los Angeles) Range, which includes the San Bernardino Mountains, and the Sierra Nevada Range. The Rocky Mountain National Park (RMNP) is also highlighted as a supplemental area. A nationwide description of emissions, concentrations, and deposition is provided in Section 3.2; a detailed characterization of nitrogen and sulfur deposition in and near the case study areas¹ is presented in Section 3.3; and the relative contributions of ambient concentrations to deposition are evaluated in Section 3.4. The deposition fields described here will be used as modeling input for the individual case study ecological modeling presented in Chapters 4 and 5.

¹ The eight case study areas are shown in Figure 2.1-1 and discussed in Chapters 4 and 5 and Appendices 4 through 7.

1 3.1 SCIENCE OVERVIEW

2 Prior to analyzing the effects of nitrogen and sulfur deposition to the environment, the
3 ambient emissions, transformations, and transport of nitrogen and sulfur in the atmosphere must
4 first be examined. As noted in Chapter 1, the terms “oxides of nitrogen” and “nitrogen oxides”
5 (NO_x) refer to all forms of oxidized nitrogen compounds, including nitric oxide (NO), nitrogen
6 dioxide (NO_2), and all other oxidized nitrogen-containing compounds transformed from NO and
7 NO_2 . Additionally, reduced forms of nitrogen (ammonia [NH_3] and ammonium ion [NH_4^+],
8 collectively termed reduced nitrogen [NH_x]) can also play an important role in the emission,
9 transformation, and deposition, and are included in this review. Much like NO_x , additional x can
10 lead to increased acidification and nutrient enrichment in ecosystems. Where possible, the
11 analyses will separate oxidized from reduced forms of nitrogen to show the impact from each
12 component, as well as the overall impact from total reactive nitrogen. This will be important for
13 the policy assessment portion of this review.

14 Sulfur oxides (SO_x) refer to all oxides of sulfur, including sulfur monoxide (SO), sulfur
15 dioxide (SO_2), sulfur trioxide (SO_3), and disulfur monoxide (S_2O); however, only SO_2 is present
16 in concentrations relevant for atmospheric chemistry and ecological exposures.

17 Deposition of nitrogen and sulfur to water and land surfaces is a function of ambient
18 concentrations of NO_x , x , and SO_x and of surface properties through complex processes involving
19 numerous meteorological parameters and dependencies. Atmospheric pollutants deposit through
20 direct contact with the surface (i.e., dry deposition), transfer into liquid precipitation (i.e., wet
21 deposition), and through interaction with fog or mist (i.e., occult deposition). Occult deposition is
22 not routinely measured and, therefore, was not taken into account for this review. Wet and dry
23 deposition are the two major mechanisms of deposition addressed here. The magnitude of wet
24 and dry deposition is related to the ambient concentrations of NO_x and SO_x through the time-,
25 location-, process-, and species-specific deposition velocity (Seinfeld and Pandis, 1998). The
26 ambient concentrations of NO_x , NH_3 , and SO_2 that contribute to nitrogen and sulfur deposition
27 are the result of emissions of these pollutants and oxidant precursor species (e.g., volatile organic
28 compounds) from anthropogenic and natural sources. The emissions-to-concentration-to-
29 deposition processes involving the chemical formation and fate of gas and particle-phase total
30 reactive nitrogen and sulfur are described in Chapter 2.6 of the *Integrated Science Assessment*
31 (*ISA*) for *Oxides of Nitrogen and Sulfur—Ecological Criteria (Final Report)* (ISA) (U.S. EPA,

1 2008b). **Figure 1.3-1** illustrates the cycle of reactive, oxidized nitrogen species in the
2 atmosphere. Emissions of NO_x lead to NO and NO_2 concentrations that can react to form other
3 nitrogen-containing oxidants. Because NO and NO_2 are only slightly soluble, they can be
4 transported over longer distances in the gas phase than more soluble pollutants. During transport,
5 NO and NO_2 can be transformed into other pollutants, such as peroxyacetyl nitrates (PAN),
6 which can provide a major source of NO_x in remote areas. NO_2 can also form gas-phase nitric
7 acid (HNO_3), which can increase the acidity of clouds, fog, and rain water and form particulate
8 nitrate that contributes to nitrogen deposition in locations distant from the NO_x emissions source
9 area. Emissions of SO_x contain SO_2 , which is oxidized in the atmosphere through a series of
10 reactions with hydroxide (OH^-), oxygen (O_2), and water (H_2O) to form sulfuric acid (H_2SO_4).
11 H_2SO_4 is also formed from SO_3 emissions within or immediately after release into the
12 atmosphere. H_2SO_4 is rapidly transformed to the aqueous phase of aerosol particles and cloud
13 droplets and can participate in the formation of new particles. The transformations of sulfur
14 compounds in the atmosphere is illustrated in **Figure 1.3-2**. Emissions of NH_3 neutralize the
15 acidity in ambient particles and form new particles through reactions with gas-phase HNO_3 to
16 form ammonium nitrate (NH_4NO_3) and with sulfate (SO_4^{2-}) to form ammonium sulfates, which
17 are significant components of nitrogen and sulfur deposition. Thus, NO_x , SO_x , and NH_3
18 emissions can not only affect atmospheric loadings of these pollutants in and near source
19 locations, but they can also affect more distant areas through chemical transformation and
20 transport.

21 **3.2 NATIONWIDE SOURCES, CONCENTRATIONS, AND** 22 **DEPOSITION OF NO_x , NH_3 , AND SO_x**

23 **3.2.1 Sources of Nitrogen and Sulfur**

24 The National Emissions Inventory (NEI) annual total emissions data for 2002 (U.S. EPA,
25 2006) are used to characterize the magnitude and spatial patterns in emissions of NO_x , NH_3 , and
26 SO_2 nationwide². The spatial resolution of these data varies by source type. Emissions from most
27 large stationary sources are represented by individual point sources (e.g., electric generating
28 units, industrial boilers). Sources that emit over broad areas are reported as county total
29 emissions (e.g., mobile sources). The national annual 2002 emissions of NO_x , NH_3 , and SO_2 by

² For the purposes of this analysis, nationwide emissions do not include emissions from Alaska or Hawaii.

1 major source category are presented in Table 2-1 of the ISA (U.S. EPA, 2008b). In total, for all
2 source categories combined, emissions of NO_x are the largest of these three pollutants at >20
3 million metric (MM) tons/yr followed by SO_x at >16 MM tons/yr. Emissions of NH₃, at >4 MM
4 tons/yr, are relatively small by comparison to emissions of NO_x and SO_x.

5 NO_x Emissions

6 The distribution of NO_x emissions across major source categories is shown in
7 **Figure 3.2-1**. Charts are provided to show emissions by major source category on a national total
8 basis, as well as for the East³ and West to reveal regional differences in source emissions
9 profiles. In addition to anthropogenic sources, there are also natural sources of NO_x, including
10 lightning, wildfires, and microbial activity in soils (U.S. EPA, 2008b, AX2, Section 2.1.2).
11 Nationally, anthropogenic sources account for the vast majority of total NO_x emissions, with
12 combustion sources as the largest contributors. Transportation-related sources (i.e., on-road,
13 nonroad, and aircraft/locomotive/marine) account for ~60% of total anthropogenic emissions of
14 NO_x, while stationary sources (e.g., electrical utilities and industrial boilers) account for most of
15 the remainder (U.S. EPA, 2008b, AX2, Table 2-1). Emissions from on-road vehicles represent
16 the major component of mobile source NO_x emissions. Approximately half the mobile source
17 emissions are contributed by diesel engines, and half are emitted by gasoline-fueled vehicles and
18 other sources (U.S. EPA, 2008b, AX2, Section 2.1.1 and Table 2-1). Nationwide, the nonroad,
19 aircraft/locomotive/marine, and non-electric generating unit (EGU) point emissions sectors each
20 contribute generally similar amounts to the overall NO_x inventory. Overall, NO_x emissions are
21 broadly split between NO and NO₂ in a ratio of 90% NO and 10% directly emitted NO₂.
22 However, this split can vary by source category, as described in Chapter 2.2.1 of the ISA (U.S.
23 EPA, 2008b).

³ In this analysis, the East is defined as all states from Texas northward to North Dakota and eastward to the East Coast of the United States. States from New Mexico northward to Montana and westward to the West Coast are considered to be part of the West.

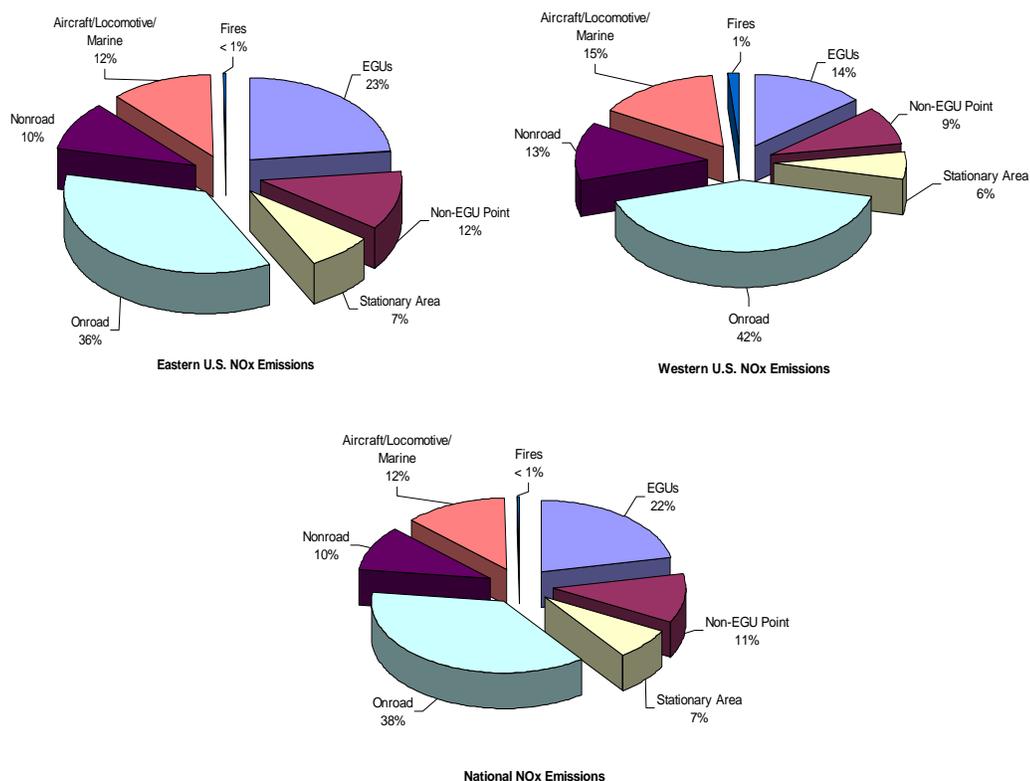


Figure 3.2-1. Annual NO_x emissions across major source categories in 2002.

Note: EGU = electric generating unit (refers to emissions from utilities).

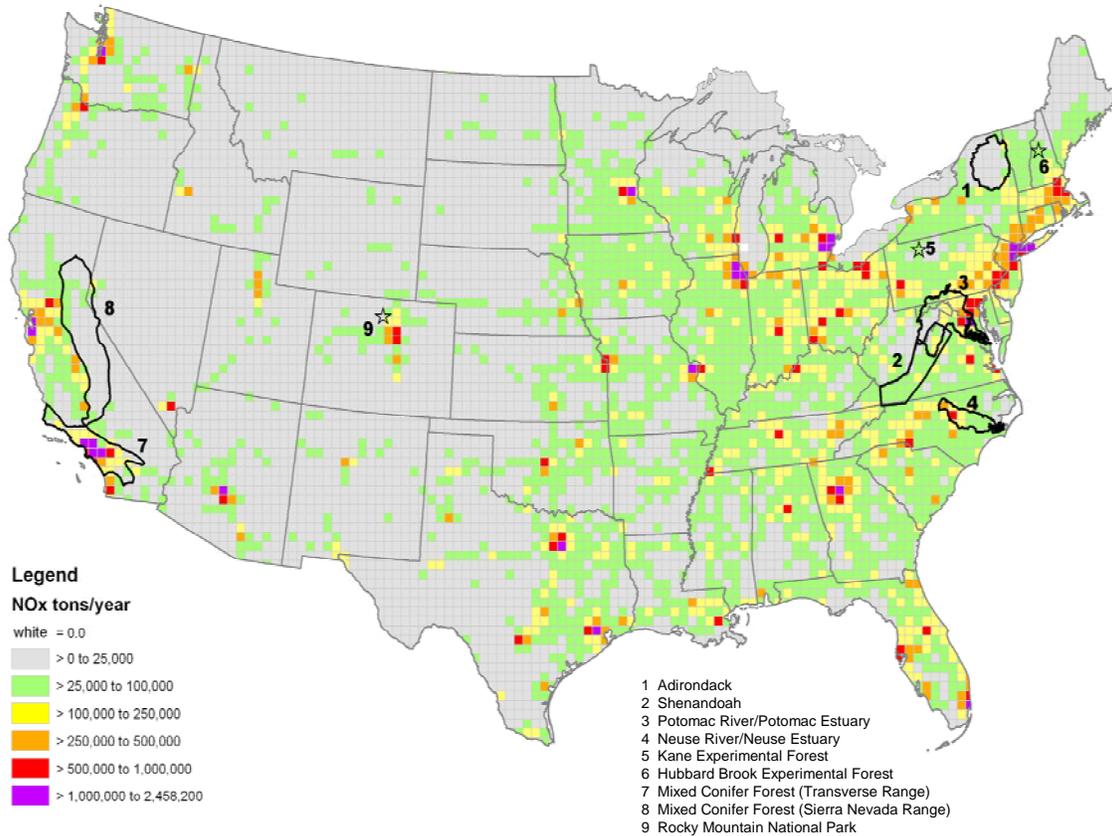
In general, NO_x emissions in the East are far greater than emissions in the West. Most of the NO_x in the West is emitted from sources in California (not shown). In the eastern and western United States, the on-road sector is the largest contributor, followed by emissions from utilities' EGUs. Although NO_x emissions from fires are a relatively small fraction of the annual total emissions in the West, fires are episodic events, and emissions can be quite high during those events.

The spatial patterns of 2002 annual NO_x emissions across the United States are shown in **Figure 3.2-2**⁴. Emissions of NO_x are concentrated in and near urban and suburban areas and along major highways. Moderate or higher levels of NO_x emissions (>100,000 tons/yr)⁵ are also

⁴ To create this map, NO_x emissions were allocated to a 36 x 36– km grid covering the United States in order to normalize for the differences in the geographic aggregation of point- and county-based emissions. The emissions are in tons per year per 36 x 36 km (1,296 km²).

⁵ Emissions are in tons per year per 36 x 36 km (1,296 km²).

1 evident in some rural areas at locations (i.e., grid cells) containing major point sources. The
 2 amount of NO_x emissions in and near each of the case study areas can be seen from this map. All
 3 of the case study areas contain or are near locations with NO_x emissions in excess of 100,000
 4 tons/yr.

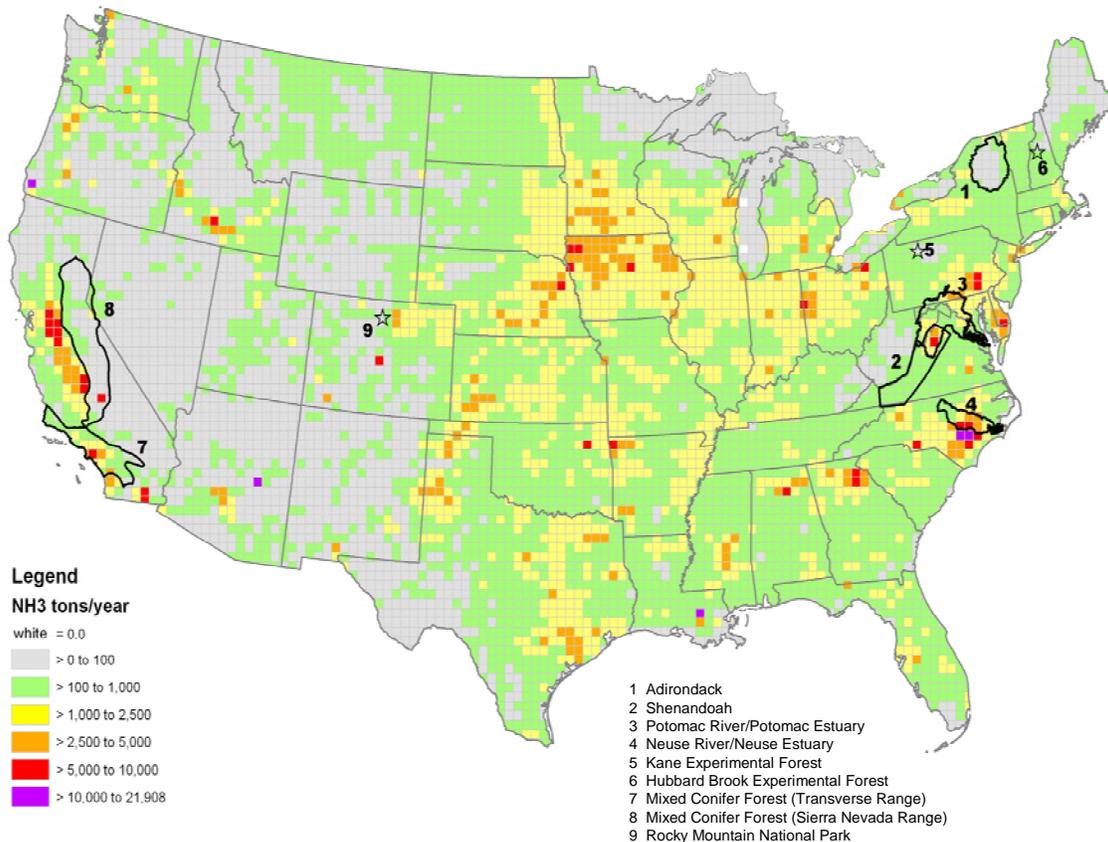


5
 6 **Figure 3.2-2.** Spatial distribution of annual total NO_x emissions (tons/yr) for 2002.

7 **NH₃ Emissions**

8 The primary anthropogenic sources of NH₃ emissions are fertilized soils and livestock.
 9 Confined animal feeding operations and other intensified agricultural production methods have
 10 resulted in greatly increased volumes of animal wastes, of which 30% to 70% may be emitted as
 11 NH₃. Motor vehicles and stationary combustion are small emitters of NH₃. Some NH₃ is emitted
 12 as a byproduct of NO_x reduction in motor vehicle catalysts. The spatial patterns of 2002 annual

1 NH₃ emissions are shown in **Figure 3.2-3**⁶. The highest emissions of NH₃ are generally found in
 2 areas of major livestock feeding and production facilities, most of which are in rural areas. In
 3 addition, NH₃ emissions exceeding 1,000 tons/yr are evident across broad areas that are likely
 4 associated with the application of fertilizer to crops. The patterns in NH₃ emissions are in
 5 contrast to the more urban-focused emissions of NO_x. The Potomac River/Potomac Estuary,
 6 Neuse River/Neuse River Estuary, Shenandoah, and Mixed Conifer Forest (in the Sierra Nevada
 7 Range and the Transverse Range) case study areas all have sources with NH₃ emissions
 8 exceeding 5,000 tons/yr. Rocky Mountain National Park is adjacent to an area with relatively
 9 high NH₃ emissions exceeding 2,500 tons/yr. The Adirondack, Hubbard Brook Experimental
 10 Forest, and Kane Experimental Forest case study areas are more distant from sources of NH₃ of
 11 this magnitude.



12
 13 **Figure 3.2-3.** Spatial distribution of annual total NH₃ emissions (tons/yr) for 2002.

⁶ Note that, because overall emissions of NH₃ are much lower than emissions of NO_x, we used a more refined set of ranges to display emissions of NH₃ compared to what was used to display emissions of NO_x.

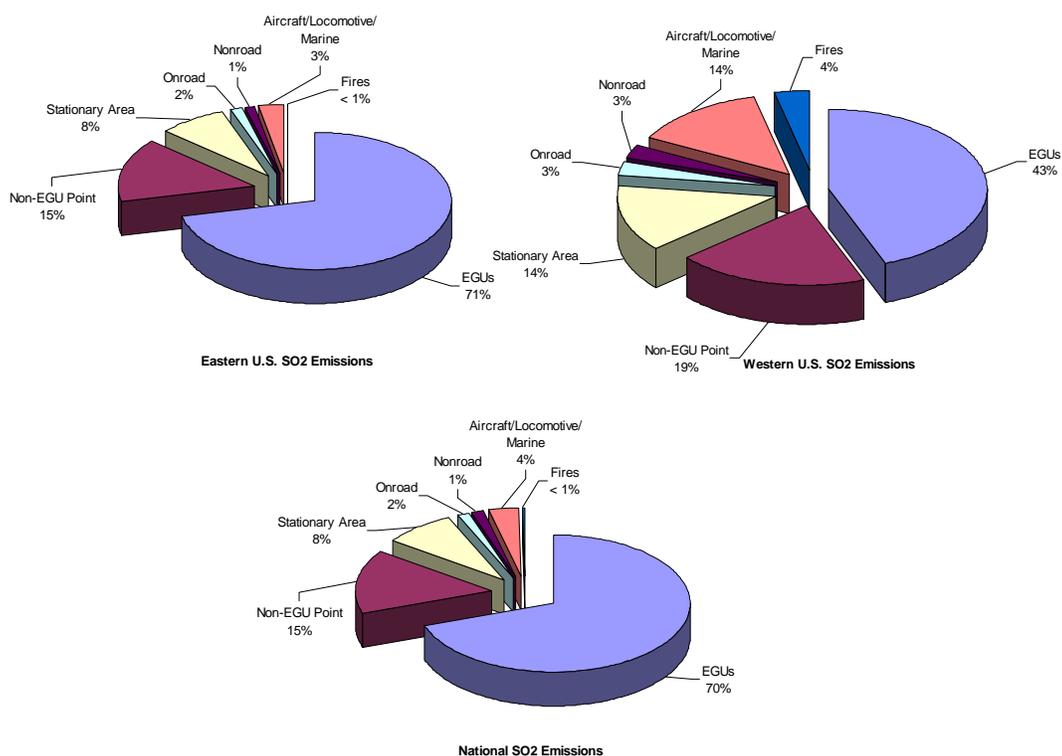
1 **SO_x Emissions**

2 The distributions of SO₂ emissions for major source categories nationally and in the East
3 and West are shown in the pie charts in **Figure 3.2-4**. Anthropogenic emissions of SO₂ in the
4 United States are mainly due to combustion of fossil fuels by electrical utilities (~70%) and non-
5 EGU sources (~15%); transportation-related sources contribute minimally (~7%). Thus, most
6 SO₂ emissions originate from point sources. Almost all the sulfur in fuel is released as volatile
7 components (SO₂ or SO₃) during combustion. The higher sulfur content of coal compared to
8 other types of fossil fuels results in higher SO₂ emissions from electrical utilities using coal as
9 fuel.

10 Similar to emissions of NO_x, emissions of SO₂ are much greater in the East than in the
11 West. The breakout of SO₂ emissions by source sector indicates that EGU emissions dominate in
12 both the East and West, but are a much greater fraction of the inventory in the East (71%)
13 compared to the West (43%). In the West, stationary area sources and non-EGU point sources
14 also have a greater contribution to SO₂ than in the East⁷.

15 The largest natural sources of SO₂ are volcanoes and wildfires. Although SO₂ constitutes
16 a relatively minor fraction (0.005% by volume) of total volcanic emissions (Holland, 1978),
17 concentrations in volcanic plumes can be range up to tens of parts per million (ppm). Sulfur is a
18 component of amino acids in vegetation and is released during combustion. Emissions of SO₂
19 from burning vegetation are generally in the range of 1% to 2% of the biomass burned (Levine et
20 al., 1999).

⁷ Note that SO₂ emissions from fires are understated in the NEI because of an error in the emissions calculations.

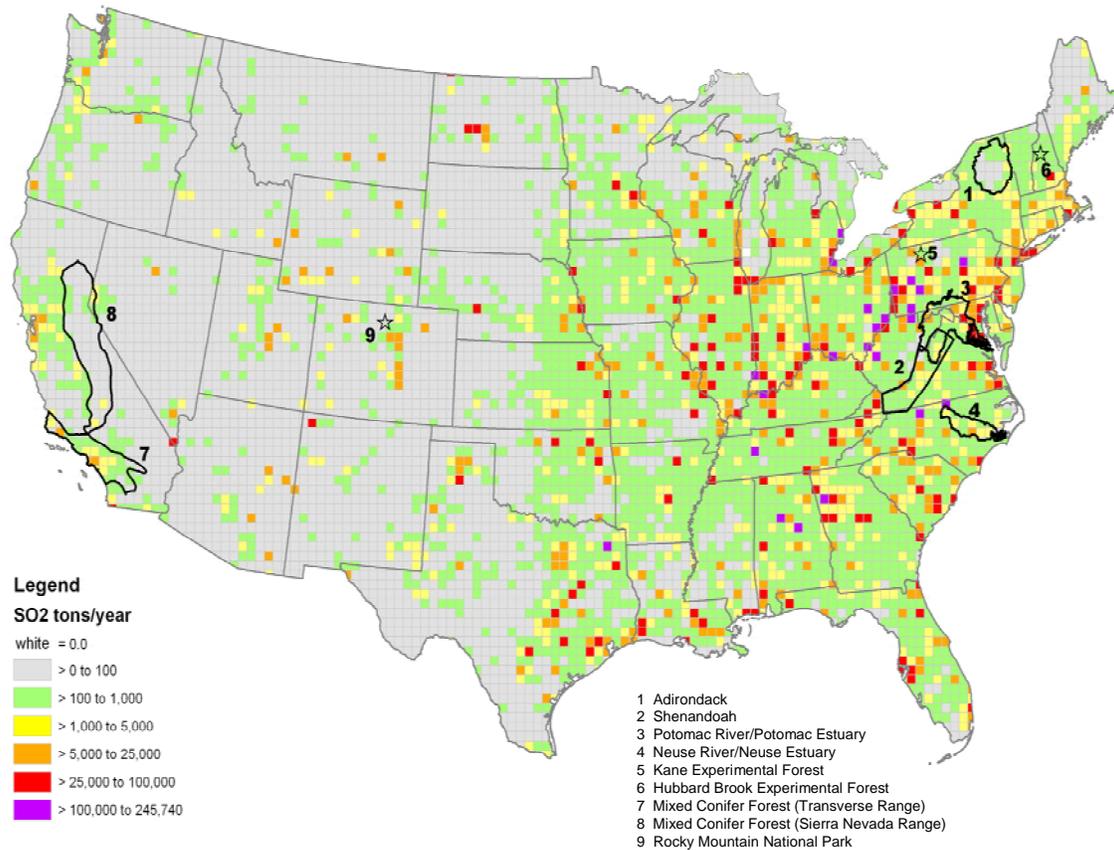


1
2 **Figure 3.2-4.** Annual SO₂ emissions across major source categories in 2002.

3 **Note:** EGUs = Electric generating unit (refers to emissions from utilities).

4 The spatial patterns of 2002 annual SO₂ emissions are shown in **Figure 3.2-5**. High SO₂
5 emissions are scattered across the East, and there are large sources in both urban are rural
6 locations. The greatest geographic concentration of SO₂ sources is in the Midwest, particularly
7 along the Ohio River, where numerous electric generating units are located. As noted above, SO₂
8 emissions in the West are much lower than in the East, with sources concentrated in urban
9 locations along with localized emissions in more rural areas associated with industrial sources
10 (e.g., smelters) and gas-field operations.

11 The Potomac River/Potomac Estuary, Neuse River/Neuse River Estuary, Shenandoah,
12 and Mixed Conifer Forest (Transverse Range portion) case study areas each contain numerous
13 locations of SO_x emissions. The Kane Experimental Forest Case Study Area and Rocky
14 Mountain National Park are relatively close to SO_x emission locations exceeding 5,000 tons/yr.
15 The Adirondack, Hubbard Brook Experimental Forest, and Mixed Conifer Forest (Sierra Nevada
16 Range portion) case study areas are more distant from SO_x sources of this magnitude.



1
2 **Figure 3.2-5.** Spatial distribution of annual total SO₂ emissions (tons/yr) for 2002.

3 **3.2.2 Nationwide Atmospheric Concentrations of NO_x and SO_x**

4 This section provides a nationwide view of the magnitude and spatial patterns in
5 atmospheric concentrations of NO_x and SO_x. Measurements of these pollutants are made at
6 numerous sampling sites comprising several routine and special study monitoring networks in
7 the United States (see Section 2.9 of the ISA [U.S. EPA, 2008b] for a comprehensive review of
8 these networks and measurement techniques). Monitoring data generally provide the most direct
9 approach to characterizing concentrations in a particular location. However, for NO_x, the lack of
10 geographic coverage and limitations in spatial representativeness of most existing sites affect the
11 extent to which these monitoring data can be used to infer NO_x concentrations in unmonitored
12 areas, particularly rural locations. As noted in the ISA (U.S. EPA, 2008b), ambient NO₂ (NO_x) is
13 normally measured at only a few locations in a given area In view of the limitations of existing

1 monitoring networks, and the large spatial gradients in NO₂ concentrations, the ISA suggests that
2 air quality model predictions might be helpful for capturing the large-scale features of NO₂
3 concentrations and could be used in conjunction with measurements to provide a more complete
4 picture of the variability of NO₂ across the United States. Monitoring data are not as spatially
5 limited for SO_x as for NO_x because SO_x measurements are also available from the Clean Air
6 Status and Trends Network (CASTNET; <http://www.epa.gov/CASTNET>), which covers rural
7 and remote locations, particularly in the eastern United States

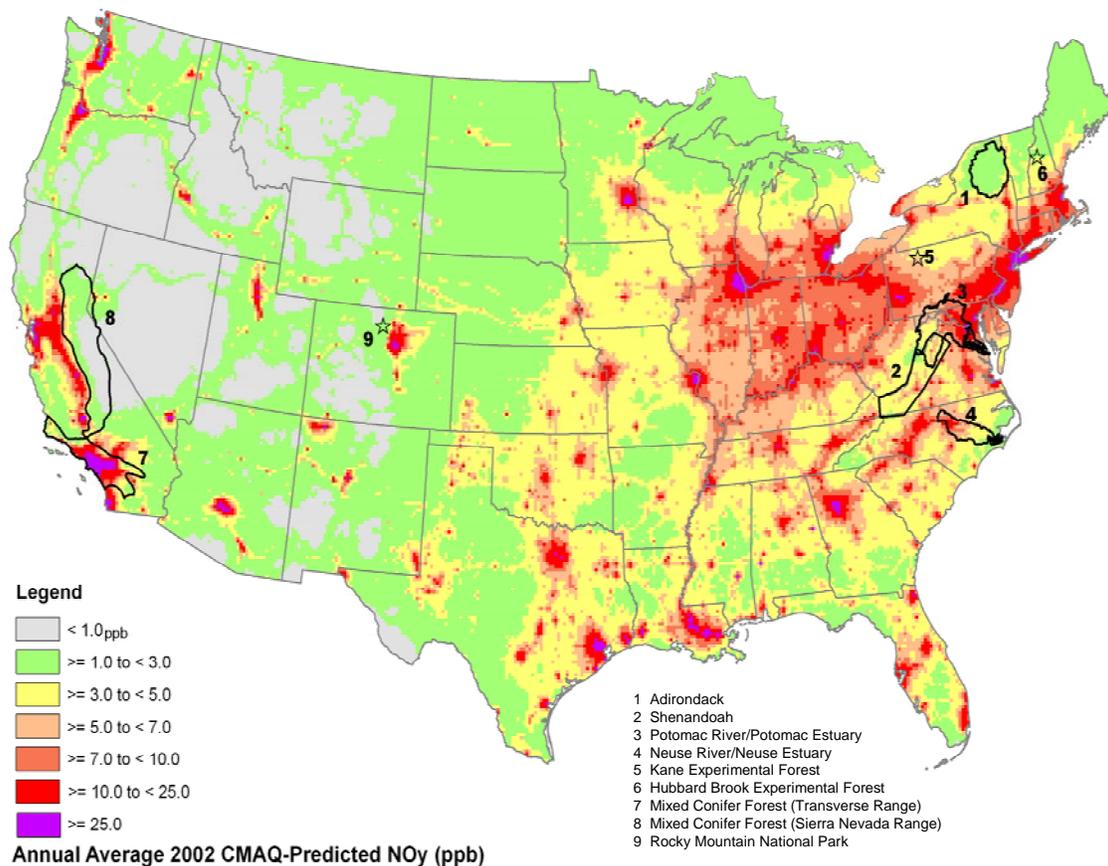
8 This analysis used measured data, along with air quality model predictions of NO_x and
9 SO₂, to characterize NO₂ and SO₂ concentrations in the United States. The air quality model
10 predictions were taken from applications of the Community Multiscale Air Quality modeling
11 system (Byun and Schere, 2006; U.S. EPA, 1999). CMAQ is a chemistry transport model that
12 treats the chemical interactions among NO_x; SO_x; other pollutants and their precursors; the
13 formation of secondary aerosols containing nitrogen, sulfur, and other species; the multi-day
14 transport of these pollutants from local to national scales; and the removal of pollutants by
15 deposition. CMAQ was used to simulate concentrations and deposition for 2002 using
16 meteorology and emissions for that year. In this application, CMAQ was run with a horizontal
17 resolution of approximately 12 × 12 km. Hourly predictions of NO_x and SO₂ were aggregated to
18 provide annual average concentration fields of these pollutants across the United States.
19 Additional information on this CMAQ application is provided in Appendix 1 of this report.

20 NO_x Concentrations

21 For the period 2003 through 2005, mean annual average NO₂ concentrations were ~15
22 parts per billion (ppb) with an interquartile range of 10 to 25 ppb and a 90th percentile value of
23 ~30 ppb, based on measurements at all monitoring sites within metropolitan statistical areas
24 (MSAs) in the United States (U.S. EPA, 2008b). Nationwide, NO₂ concentrations have been
25 trending downward, with an overall 30% decrease in concentrations from 1990 to 2006 (U.S.
26 EPA, 2008b) as a result of various federal and state NO_x emissions-control programs.

27 The spatial field of model-predicted 2002 annual average NO_x concentrations is shown in
28 **Figure 3.2-6**. The patterns in NO_x concentrations generally mirror the patterns of NO_x emissions
29 shown in **Figure 3.2-2**. For the most part, highest concentrations are predicted in the core
30 portions of urban areas with a relatively large drop in concentrations with distance from the
31 location of peak values. The spatial gradients from urban and rural areas appear to be greater in
32 the West compared to those in the East. In the West, NO_x concentrations outside source areas

1 drop off rapidly to below 3 ppb. Annual average concentrations of NO_x are predicted to exceed 3
 2 ppb in rural areas within broad portions of the East. The highest rural concentrations in the East
 3 extend across portions of the Midwest, Pennsylvania, and along the Northeast Corridor. Annual
 4 average NO_x concentrations exceeding 10 ppb are predicted in portions of the Potomac
 5 River/Potomac Estuary, Neuse River/Neuse River Estuary, and Mixed Conifer Forest
 6 (Transverse Range portion) case study areas. The Kane Experimental Forest Case Study Area is
 7 within the area of regionally high NO_x that extends across Pennsylvania. The other case study
 8 areas (Adirondack, Hubbard Brook Experimental Forest, and Mixed Conifer Forest [Sierra
 9 Nevada Range portion]) as well as the Rocky Mountains are predicted to have annual average
 10 NO_x concentrations of ~3 ppb or less.



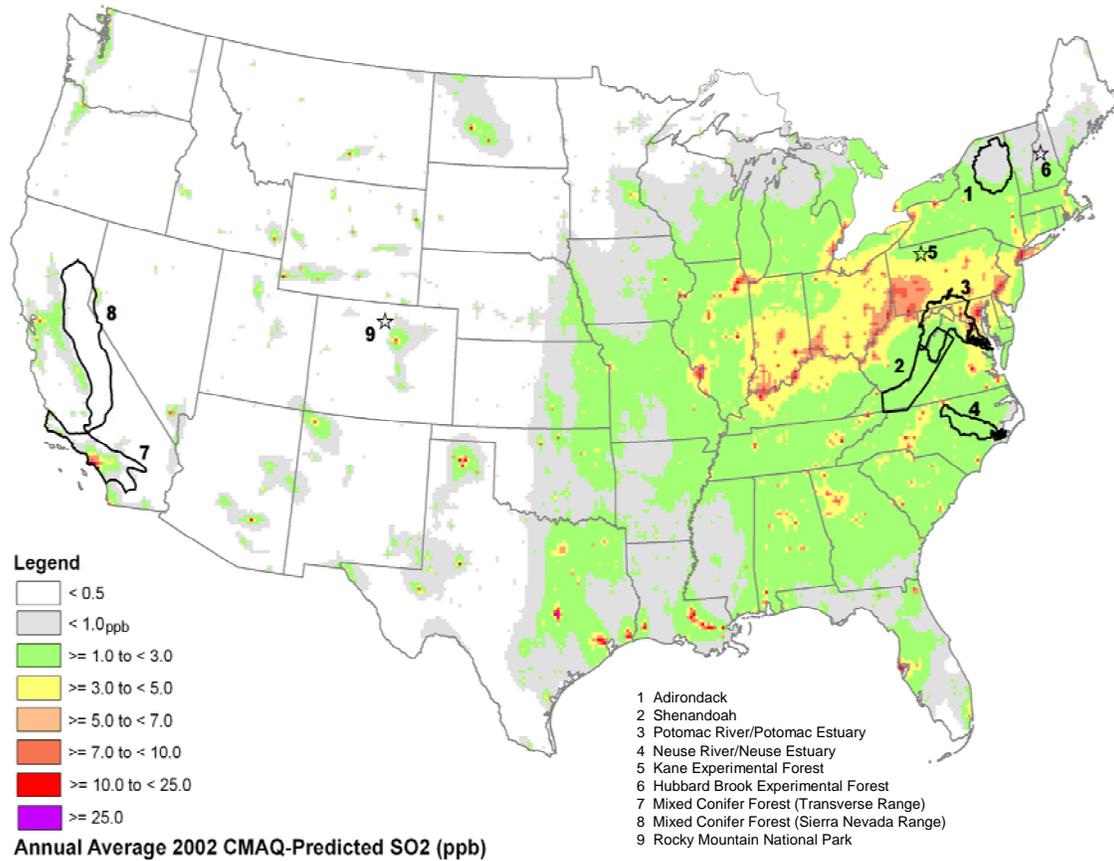
11
 12 **Figure 3.2-6.** Model-predicted annual average NO_y concentrations (ppb) for 2002.

1 **SO₂ Concentrations**

2 Measured annual average SO₂ concentrations for the period 2003 through 2005 are
3 presented in Table 2-23 of the ISA (U.S. EPA, 2008b). SO₂ concentrations aggregated across
4 urban sites and nonurban sites were generally very low at ~4 ppb. Interquartile concentrations
5 were in the range of 1 to 6 ppb for urban sites and 1 to 5 ppb for nonurban sites. Urban and non-
6 urban concentrations at the 90th percentile were 8 ppb. In an analysis of 11 cities, sites with the
7 highest annual mean SO₂ concentrations were in Steubenville, OH (8.6 to 14 ppb), and
8 Pittsburgh, PA (6.8 to 12 ppb) (U.S. EPA, 2008b). Both of these cities are in areas with very high
9 SO₂ emissions from electric generating units. At suburban and rural CASTNET sites, annual
10 average SO₂ concentrations in 2007 were much higher by far at sites in the East compared to the
11 West (U.S. EPA, 2008a). In the East, the highest concentrations were measured across the
12 Midwest, Southeast, and mid-Atlantic states. Relatively low concentrations were measured
13 across New England.

14 The 2002 annual average model-predicted SO₂ concentration fields are shown in
15 **Figure 3.2-7**. The model predictions are generally consistent with the magnitude and spatial
16 patterns of concentrations from measured data. Peak predicted concentrations, exceeding 10.0
17 ppb, coincide with the location of highest emissions (see **Figure 3.2-5**), with large decreases in
18 concentrations with distance from sources. In the East, the localized peak concentrations are
19 within a broad area of concentrations exceeding 1.0 ppb. SO₂ predictions exceed 3.0 ppb in
20 portions of the Midwest, across Pennsylvania, and into the mid-Atlantic states and decline to
21 <0.5 ppb in northern Maine. In the West, SO₂ predictions are much lower than in the East and
22 are generally <0.5 ppb, except in the vicinity of sources of SO₂.

23 The Potomac River/Potomac Estuary Case Study Area has the highest SO₂ predictions
24 among the six case study areas in the East. The portion of the Potomac River/Potomac Estuary
25 Case Study Area in western Virginia is predicted to have concentrations in the range of 1 to 3
26 ppb, which increases to 3 to 5 ppb in Maryland. SO₂ concentrations in the Kane Experimental
27 Forest, Shenandoah, and Neuse River/Neuse River Estuary case study areas are in the range of
28 1.0 to 3.0 ppb, with some locations having up to 3.0 to 5.0 ppb. The Adirondack, Hubbard Brook
29 Experimental Forest, Mixed Conifer Forest (Sierra Nevada Range portion) case study areas, as
30 well as the Rocky Mountains, all have predicted SO₂ concentrations of <1.0 ppb. The portion of
31 Mixed Conifer Forest (Transverse Range portion) Case Study Area near the city of Los Angeles
32 has predictions exceeding 10.0 ppb.



1
2 **Figure 3.2-7.** Model-predicted annual average SO₂ concentrations (ppb) for 2002.

3 **3.2.3 Nationwide Deposition of Nitrogen and Sulfur**

4 As noted in Section 3.1 of this report, gaseous and particulate deposition of nitrogen and
5 sulfur species to land and water surfaces occurs through both dry deposition and wet deposition
6 processes. Additionally, nitrogen deposition is composed of both oxidized and reduced forms of
7 total reactive nitrogen. The nationwide analysis of deposition examined the magnitude and
8 spatial patterns of total sulfur deposition, total nitrogen deposition, and the oxidized and reduced
9 forms of total reactive nitrogen. The analysis of current levels and trends in nitrogen and sulfur
10 deposition is based in part on measured data as described in Section 2.10 of the ISA (U.S. EPA,
11 2008b). A combination of measured data and model predictions to glean additional information
12 on the magnitude and spatial patterns in deposition across the United States was also used.

1 **Approach for Assimilating Measured Data and Model Predictions**

2 To create spatial fields of deposition, wet deposition measurements from the National
3 Atmospheric Deposition Program (NADP) National Trends Network
4 (<http://nadp.sws.uiuc.edu/nadpoverview.asp>) were used. Estimates of dry deposition are
5 available from the CASTNET network (<http://www.epa.gov/castnet/>) (Clarke et al., 1997), but
6 these data are calculated based on an “inferential model” involving measured air concentrations
7 coupled with species- and location- dependent deposition velocities that reflect local land use
8 and meteorological conditions at each monitoring site (EPA, 2008b). These dry deposition
9 estimates may not be representative of dry deposition fluxes in unmonitored areas where land use
10 or meteorological conditions are different from those at monitoring sites. Therefore, for the
11 nationwide assessment of deposition, dry deposition predictions from the 2002 CMAQ model
12 simulation was used because the model has information about meteorology and land use in each
13 grid cell of the domain not merely where the monitors are positioned. Thus annual total 2002 wet
14 deposition from NADP measurements, coupled with the 2002 model-predicted dry deposition
15 from CMAQ, were used.

16 NADP data are collected at several hundred point locations across the contiguous United
17 States. From these points, analysts at the NADP generated continuous surfaces at a 2.5-km grid
18 cell resolution by using an inverse distance weighted (IDW) algorithm available at
19 <http://www.epa.gov/monitor/programs/nadpntn.html>. Wet deposition of sulfur was calculated
20 from deposition measurements of sulfate (SO_4^{2-}). Oxidized nitrogen wet deposition was
21 calculated from measured nitrate (NO_3^-) deposition, and reduced nitrogen deposition was
22 calculated from deposition of ammonium (NH_4^+).

23 The CMAQ data were generated at a 12-km grid cell size and consisted of many
24 estimated deposition values, including total dry sulfur, total dry nitrogen, total dry oxidized
25 nitrogen, and total dry reduced nitrogen. The oxidized nitrogen species from CMAQ are NO_3^- ,
26 HNO_3 , NO , NO_2 , dinitrogen pentoxide (N_2O_5), PAN, HONO, and organic nitrates (NTR), while
27 the reduced nitrogen species are NH_3 and NH_4^+ . Both the measured and modeled datasets
28 provided deposition values in kg/ha/yr. The NADP data were at a finer spatial resolution, and in
29 order to add the two gridded datasets together, the finer NADP dataset was resampled up to the
30 12-km scale of the CMAQ data. Once both datasets were at the same spatial resolution, the wet
31 and dry deposition values for each component (e.g., oxidized nitrogen) were added together on a
32 grid-cell by grid-cell basis to provide spatial fields of estimated annual total (i.e., wet plus dry)

1 deposition across the United States. The combined measured plus modeled deposition fields
2 were also used as input for the individual case study ecological modeling described in Chapters 4
3 and 5 and Appendices 4 through 7 of this report.

4 **Nitrogen Deposition**

5 As noted in the ISA, increasing trends in urbanization, agricultural intensity, and
6 industrial expansion during the previous 100 years have produced a nearly ten-fold increase in
7 atmospherically deposited nitrogen (U.S. EPA, 2008b). Increased deposition of reduced nitrogen
8 in the United States, measured as NH_4^+ deposition, correlates well with the local and regional
9 increases in agricultural intensity. Although aggregate nitrogen deposition trends based on a
10 sample of 34 NADP sites in the East show an overall decline from deposition levels in 1990,
11 more recent trends beginning in the late 1990s have been less consistent (U.S. EPA, 2008b;
12 Sickles and Shadwick, 2007a).

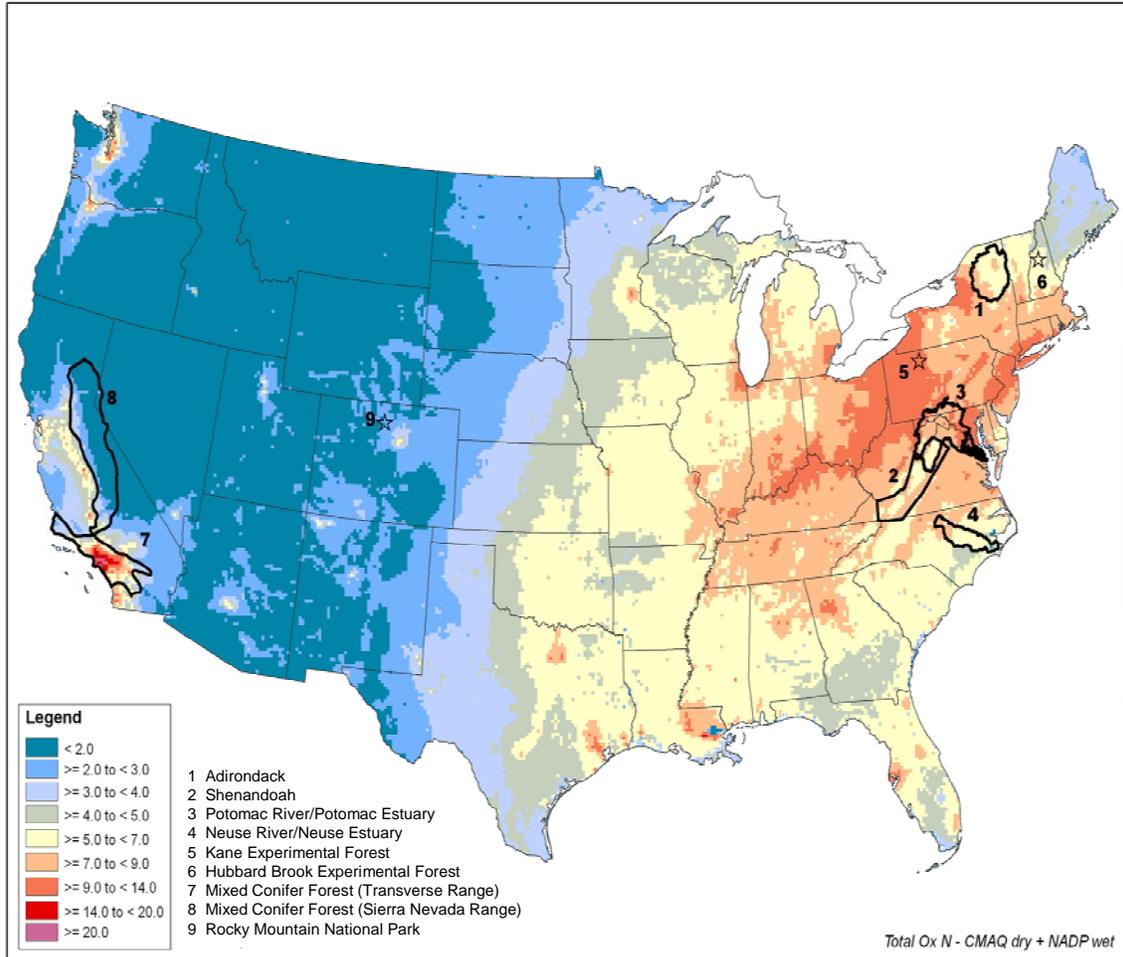
13 From 2004 to 2006, measured deposition was greatest in the Ohio River Valley,
14 specifically in Indiana and Ohio, where there were values as high as 9.2 and 9.6 kg N/ha/yr,
15 respectively. Nitrogen deposition was lower at sites in other parts of the East, including Florida
16 and in northern New England, where nitrogen deposition was 4.0 kg N/ha/yr or less. The greatest
17 deposition in the central United States occurred in Kansas and Oklahoma, which reported 7.0 and
18 6.5 kg N/ha/yr, respectively. Measured nitrogen deposition levels were much lower in the West
19 where values ranged from ~1.0 to 3.0 kg N/ha/yr. The highest deposition in the West (~4.0 to 5.0
20 kg N/ha/yr) was measured at sites near Los Angeles, CA. In most areas of the country, measured
21 wet deposition dominates dry deposition in terms of the contribution to total deposition. The
22 extent of wet versus dry deposition varies regionally to some extent because some western sites
23 have more similar or higher rates of dry versus wet deposition than the more humid sites in the
24 East.

25 The spatial fields of oxidized nitrogen deposition, reduced nitrogen deposition, and total
26 reactive nitrogen deposition across the United States for 2002 are shown in **Figures 3.2-8, 3.2-9,**
27 **and 3.2-10**, respectively. In general, on a regional basis, both forms of nitrogen deposition are
28 much higher in the East compared to the West. Within the eastern United States, there is a broad
29 area with oxidized nitrogen deposition of 5.5 kg N/ha/yr or greater that extends from Louisiana
30 northeastward across portions of the Southeast and Midwest to the mid-Atlantic region and most
31 of New England. This area of elevated oxidized nitrogen deposition roughly corresponds to the
32 areas with model-predicted NO_x concentrations of 3.0 ppb or greater and, in general, where NO_x

1 emissions are regionally highest. Oxidized nitrogen deposition levels of 7.5 kg N/ha/yr or greater
2 are evident in and near NO_x source areas and within much of a multistate area from Tennessee
3 northeastward to central New England. In the West, oxidized nitrogen deposition is 1.5 kg
4 N/ha/yr or less across most of the region, except in urban areas, where NO_x emissions are
5 highest.

6 As shown in **Figure 3.2-9**, the geographic patterns in reduced nitrogen deposition,
7 indicate that the areas of high reduced nitrogen deposition in both the East and West generally
8 correspond to areas of high NH₃ emissions in each region (see **Figure 3.2-3**). In the East,
9 deposition of reduced nitrogen of 3.5 kg N/ha/yr or greater is seen from central Texas, across the
10 eastern Great Plains and the Midwest, to western Pennsylvania and western New York.
11 Elsewhere in the East, high levels of reduced nitrogen deposition are found in and near areas of
12 livestock/swine/poultry operations. Between these areas of elevated deposition, reduced nitrogen
13 deposition levels are generally in the range of 1.5 to 3.5 kg N/ha/yr. In the West, reduced
14 nitrogen deposition is <1.5 kg N/ha/yr, except near NH₃ emissions source areas, especially the
15 Central Valley of California.

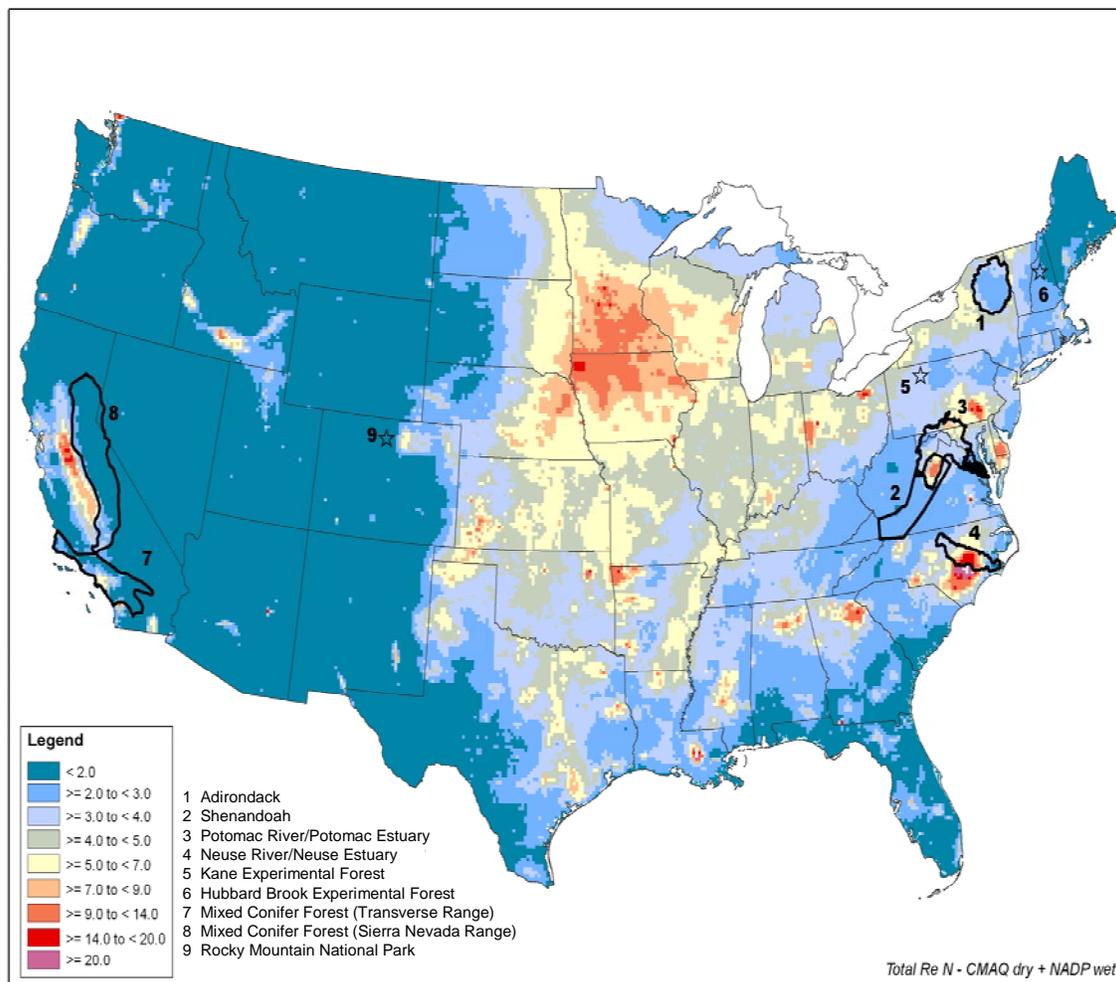
16 The spatial patterns of total reactive nitrogen deposition in **Figure 3.2-10** reflect the
17 combination of the deposition from the reduced and oxidized nitrogen components. Much of the
18 East has total nitrogen deposition of 9 to 12 kg N/ha/yr. Higher amounts of 12 kg N/ha/yr or
19 greater cover large portions of the Midwest and Northeast, as well as in or near sources of NO_x
20 and/or NH₃ emissions in other parts of the East. In the West, total nitrogen deposition is highest
21 in and near NO_x and NH₃ source areas, particularly those in California, where deposition exceeds
22 18 kg N/ha/yr. In most rural or remote portions of the West, total nitrogen deposition is generally
23 <3 kg N/ha/yr.



1

2

Figure 3.2-8. Total wet plus dry oxidized nitrogen deposition (kg N/ha/yr) in 2002.



1
2

Figure 3.2-9. Total wet plus dry reduced nitrogen deposition (kg N/ha/yr) in 2002.

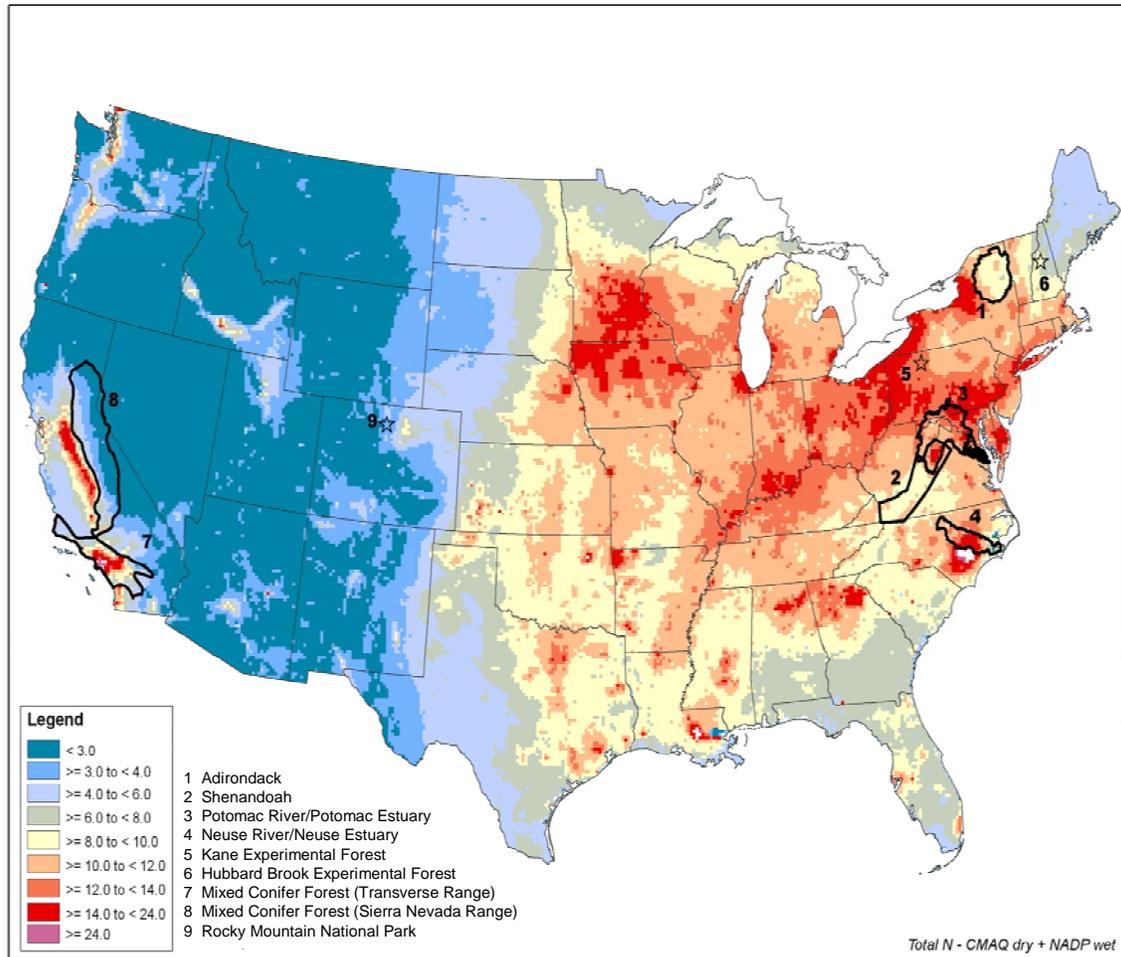


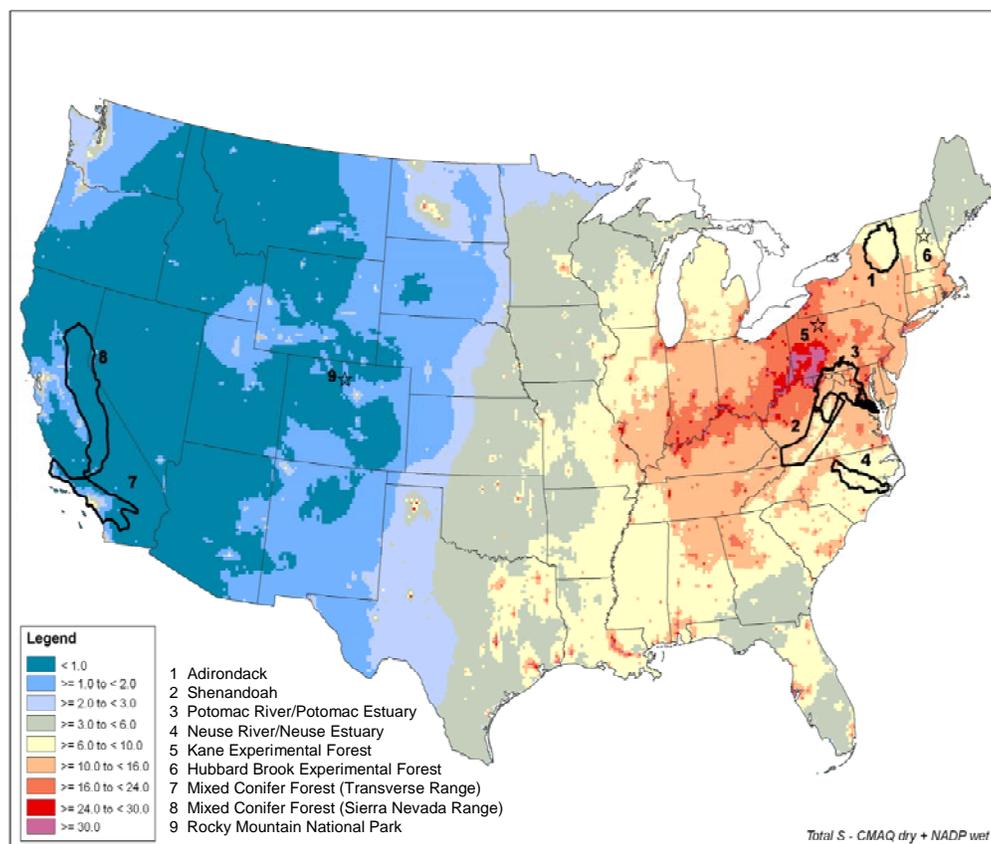
Figure 3.2-10. Total reactive nitrogen deposition (kg N/ha/yr) in 2002.

Sulfur Deposition

Annual average measured sulfur deposition during 2004 to 2006 was highest in the Ohio River Valley. In this region, measured sulfur deposition was 21.3 kg S/ha/yr at one monitoring site, and most sites reported 3-year averages >10.0 kg S/ha/yr (U.S. EPA, 2008b). Total sulfur deposition measured in the West was relatively low, and generally <2.0 kg S/ha/yr, with many sites measuring <1.0 kg S/ha/yr. The primary form of sulfur deposited is wet SO_4^{2-} . Smaller contributions to deposition are made by dry SO_2 and dry SO_4^{2-} .

The spatial fields of sulfur across the United States for 2002 are shown in **Figure 3.2-11**. As with the deposition of nitrogen species, sulfur deposition is much higher in the East than the West. Sulfur deposition across most of the West is <3.0 kg S/ha/yr. In the East, high levels of deposition exceeding 18 kg S/ha/yr are seen in the immediate vicinity of isolated major sources, as well as in and near areas having a high concentration of SO_2 sources. This is particularly

1 notable along the Ohio River Valley, extending across Pennsylvania. The areas of highest
 2 deposition are within a broad area of sulfur deposition in the range of 6 to 12 kg S/ha/yr, which
 3 covers much of the East.



4
 5 **Figure 3.2-11.** Total wet and dry sulfur deposition (kg S/ha/yr) in 2002.

6 **3.2.4 Policy-Relevant Background Concentrations**

7 Policy-relevant background concentrations are those concentrations that would occur in
 8 the United States in the absence of anthropogenic emissions in the continental North America
 9 (i.e., United States, Canada, Mexico). The analyses for the Risk and Exposure Assessment
 10 examined the contribution of total reactive nitrogen and sulfur above the policy-relevant
 11 background concentrations.

12 For NO₂, policy-relevant background concentrations are <300 parts per trillion (ppt) over
 13 most of the continental United States and <100 ppt in the eastern United States on an annual
 14 average basis (U.S. EPA, 2008b). In contrast to the levels associated with policy relevant
 15 background concentrations, 24-hour ambient NO₂ concentrations in urban areas near monitoring
 16 locations averaged <20 ppb and have a 99 percentile value of <50 ppb. Annual average NO₂

1 concentrations over the continental United States are <5 ppb for nearly all urban, rural, and
2 remote sites. According to the ISA (U.S. EPA, 2008b), background SO₂ concentrations are <10
3 ppt throughout most of the continental United States, except in areas of the Pacific Northwest,
4 where natural SO₂ sources are particularly strong because of volcanic activity. Maximum policy-
5 relevant background SO₂ concentrations are 30 ppt. In general, policy-relevant background
6 concentrations of SO₂ contribute <1% of current concentrations, except in the Pacific Northwest,
7 where policy-relevant background concentrations can contribute up to 80% (U.S. EPA, 2008b).

8 **3.2.5 Nonambient Loadings of Nitrogen and Sulfur**

9 Not all loadings of nitrogen and sulfur compounds to ecosystems are due to atmospheric
10 deposition. Other inputs, such as runoff from agricultural soils to waterbodies and point-source
11 discharges, also contribute to acidification and nutrient enrichment. This assessment examines
12 the atmospheric contribution due to total reactive nitrogen and sulfur, recognizing that some
13 systems may be solely impacted by atmospheric deposition, while effects in other systems might
14 be largely due to nonatmospheric sources. This source distinction will play an important role in
15 the standard-setting process.

16 **3.3 SPATIAL AND TEMPORAL CHARACTERIZATION OF** 17 **DEPOSITION FOR CASE STUDY AREAS**

18 **3.3.1 Purpose and Intent**

19 The purpose of this section is to describe the spatial and temporal patterns of total
20 reactive nitrogen and sulfur deposition for the eight case study areas and the Rocky Mountain
21 National Park supplemental study area. These areas are shown on the map in **Figure 2.1-1**. This
22 analysis focused on the magnitude, spatial gradients, and the intra-annual (i.e., seasonal) and
23 inter-annual (i.e., between 2002–2005) variation in nitrogen and sulfur deposition for each of
24 these case study areas. In addition to improving the overall understanding of the spatial and
25 temporal behavior of nitrogen and sulfur deposition, the results and findings of this analysis are
26 intended to provide information on the case study areas about (1) the relative portion of total
27 nitrogen deposition that is in the form of oxidized versus reduced nitrogen, and
28 (2) the relative amounts of wet versus dry deposition of nitrogen and sulfur.

1 These analyses are intended to aid in understanding the characteristic patterns of
2 deposition in the case study areas and their current contribution to negative ecological effects. It
3 is beyond the scope of this analysis to fully explain the characteristics revealed by the modeled
4 and measured deposition and concentrations. Further exploration of these relationships and
5 interactions should be the subject of future research efforts.

6 **3.3.2 Data and Analytical Techniques**

7 As previously discussed, both measured data and model predictions for the analyses were
8 used in this assessment. The measured data include wet deposition of nitrogen and sulfur, as
9 calculated from NO_3^- , NH_4^+ , and SO_4^{2-} wet deposition samples collected at NADP sites during
10 the period 2002 through 2005. These wet deposition data are available as annual totals for each
11 of the years 2002 through 2005 as spatial fields of gridded data at 12×12 km resolution for the
12 continental United States. The CMAQ⁸ model predictions include wet and dry deposition of
13 nitrogen and sulfur from applications of CMAQ over this same time period. The hourly model
14 predictions were aggregated to seasonal and annual time periods, as needed, for this assessment.

15 For 2002, CMAQ predictions were at a resolution of 12 km for the continental United
16 States⁹. These 2002 model predictions are based on model runs with CMAQ v4.6. The dry
17 deposition predictions for 2002 from CMAQ v4.6 were coupled with the 2002 NADP wet
18 deposition data to provide annual total reactive nitrogen and annual total sulfur deposition for
19 input to the aquatic and terrestrial ecosystem modeling analyses described in Chapters 4 and 5 of
20 this report. In October 2008, the EPA Office of Research and Development (ORD) released an
21 updated version of CMAQ (CMAQ v4.7) and an updated version of CMAQ's meteorological
22 preprocessor (MCIPv3.4)¹⁰. Recently, the EPA ORD used the updated versions of CMAQ and
23 Meteorology-Chemistry Interface Processor (MCIP) to remodel 2002 deposition and to model
24 2003, 2004, and 2005 deposition . These 2002 through 2005 CMAQ runs were performed at 12-
25 km resolution for the East¹¹ and at 36-km resolution for the West. This Risk and Exposure
26 Assessment uses both sets of CMAQ runs. The CMAQ v4.6 2002 predictions are used in the
27 analyses to characterize the magnitude, relative amounts, and spatial gradients in deposition

⁸ The CMAQ applications are described in detail in Appendix 1 of this report.

⁹ The CMAQ modeling domains are shown in Figure 1 of Appendix 1 of this report.

¹⁰ The scientific updates in CMAQ v4.7 and MCIP v3.4 can be found at the following web links:

http://www.cmascenter.org/help/model_docs/cmaq/4.7/RELEASE_NOTES.txt

http://www.cmascenter.org/help/model_docs/mcip/3.4/ReleaseNotes

¹¹ The 99° west meridian to separate the eastern and western United States was used in this assessment.

1 within each case study area, as well as to examine the seasonal variability in deposition for 2002.
2 The predictions for 2002 through 2005 from CMAQv4.7 were used as a consistent set of
3 estimates to assess inter-annual variability in deposition and to determine whether the magnitude
4 and relative amounts of deposition in 2002 are representative of conditions over the longer-term,
5 4-year time period. The differences in annual total reactive nitrogen between the two sets of 2002
6 data are 0.5 kg N/ha/yr or less for most of the case study areas and a largest difference of 0.7 kg
7 N/ha/yr. For sulfur deposition, the differences in the two 2002 data sets are 0.5 kg sulfur or less
8 for five of the eight case study areas and between 0.6 kg sulfur and 1 kg sulfur for the other four
9 areas. A comparison of the two sets of 2002 CMAQ predictions is presented as part of the
10 discussion on uncertainties in Section 3.5 of this report.

11 In general, the case study analyses rely upon a combination of NADP-measured wet
12 deposition and CMAQ (v4.6 or v4.7) dry deposition, with one exception. CMAQ predictions of
13 both wet and dry deposition were used in the analysis of seasonal variations because gridded wet
14 deposition data from NADP are not available at a subannual temporal resolution.

15 **Spatial Allocation of Gridded Data to Case Study Areas**

16 The gridded measured and modeled data were linked to the case study areas using several
17 geographic information systems (GIS)–based techniques that differ depending on the geographic
18 definition of each area, as follows. The Potomac River/Potomac Estuary Case Study Area and
19 Neuse River/Neuse River Estuary Case Study Area include contiguous watersheds that are
20 defined in terms of 8-digit Hydrologic Unit Codes¹² (HUCs). For these two areas, GIS was used
21 to calculate the spatially weighted average deposition for each of these areas as a whole. The
22 Adirondack Case Study Area includes individual noncontiguous watersheds¹³ that contain the
23 lakes/ponds selected for ecological modeling as part of the aquatic acidification analysis (see
24 Chapter 4 of this report). Similarly, the Shenandoah Case Study Area includes those
25 watersheds¹⁴ containing the streams selected for ecological modeling. For the Adirondack and
26 Shenandoah case study areas, individual grid cells were linked to each watershed if any part of
27 the grid cell touched a portion of a watershed in the area. The Hubbard Brook Experimental

http://www.cmascenter.org/help/model_docs/cmaq/4.7/RELEASE_NOTES.txt

http://www.cmascenter.org/help/model_docs/mcip/3.4/ReleaseNotes

¹²These codes are used to identify the drainage basins within the United States. See

<http://imnh.isu.edu/digitalatlas/hydr/huc/huctxt.htm> for additional information on HUCs.

¹³ The Adirondack watersheds are defined by 10-digit HUCs.

¹⁴ The Shenandoah watersheds are defined by 12-digit HUCs.

1 Forest, Kane Experimental Forest, and Mixed Conifer Forest (Transverse Range and Sierra
2 Nevada Range) case study areas, as well as the Rocky Mountain National Park, do not contain
3 finer geographic elements. For these areas, GIS was used to calculate the spatially weighted
4 average deposition for each area as a whole.

5 3.3.3 Characterization of Deposition in Case Study Areas

6 The characterizations of nitrogen and sulfur deposition for each case study area are
7 discussed in this section as follows:

- 8 ■ Overall area-wide magnitude of deposition in 2002
- 9 ■ Variation in annual total deposition between 2002 through 2005
- 10 ■ Relative amount of wet and dry, oxidized, and reduced nitrogen to total reactive nitrogen
11 deposition and wet and dry to total sulfur deposition in 2002
- 12 ■ Geographic variations in annual deposition for 2002 within and near the case study areas
- 13 ■ Seasonal variations in each component of deposition for 2002.

14 The table and figures that provide and display the data used for this analysis are identified
15 below. For ease of reference, the table and figures are provided at the end of each subsection.

16 The modeled plus measured annual total reactive nitrogen and sulfur depositions for 2002
17 for each case study area, as a whole, are presented in **Table 3.3-1**. The inter-annual variations in
18 total reactive nitrogen deposition from 2002 through 2005 are shown in **Figures 3.3-1(a and b)**
19 for each case study area in the East and West. The relative amounts of oxidized versus reduced
20 nitrogen deposition for each case study area in 2002 are shown in **Figure 3.3-2**. The relative
21 amounts of wet and dry, oxidized, and reduced nitrogen deposition for 2002 are shown in
22 **Figures 3.3-3(a-i)**. The spatial patterns in annual nitrogen depositions for 2002 are shown in
23 **Figures 3.3-4(a-e)** for the East and in **Figures 3.3-5(a-c)** for the West. The seasonal variations in
24 total reactive nitrogen deposition for each case study area are shown in **Figures 3.3-6(a-i)**. The
25 seasonal data are presented in terms of the percentage of annual deposition that occurs in each
26 season¹⁵. For wet and dry, oxidized and reduced nitrogen deposition seasonal variations are

¹⁵ For the purposes of this analysis, data for December, January, and February 2002 were included in “winter”; data for March, April, and May 2002 were included in “spring”; data for June, July, and August 2002 were included in “summer”; and data for September, October, and November 2002 were included in “fall.” Thus, data for December 2002 were included with data for January and February of this same year.

1 shown in **Figures 3.3-7(a-i)**, along with the seasonal variation in precipitation. Seasonal patterns
2 of NH₃ emissions are shown in **Figure 3.3-8**.

3 The annual total sulfur deposition from 2002 through 2005 is shown in **Figures 3.3-9**
4 **(a and b)** for each case study area in the East and West. The relative amounts of wet and dry
5 sulfur deposition in 2002 and, on average, for the period 2002 through 2005 are shown in
6 **Figures 3.3-10 and 3.3-11**. The spatial patterns in annual sulfur deposition for 2002 are shown in
7 **Figures 3.3-12(a-c)** for the East and in **Figure 3.3-13** for the West. The seasonal variation in
8 total sulfur deposition for each case study area is shown in **Figures 3.3-14(a-i)**. Wet and dry
9 sulfur deposition seasonal variations are shown in **Figures 3.3-15(a-i)**.

10 **3.3.3.1 Magnitude of Total Reactive Nitrogen Deposition in 2002 and Analysis of** 11 **Inter-annual Variability**

12 The amount of total reactive nitrogen deposition in 2002 varies among the case study
13 areas (see **Table 3.3-1**). In the East, total reactive nitrogen deposition ranges from 8 kg N/ha/yr
14 for the Hubbard Brook Experimental Forest Case Study Area up to 14 kg N/ha/yr for the Neuse
15 River/Neuse River Estuary Case Study Area. Total reactive nitrogen deposition in 2002 is also
16 high in the Transverse Range portion of the Mixed Conifer Forest Case Study Area (10 kg
17 N/ha/yr), which reflects the high levels of NO_x emissions in and around the Los Angeles urban
18 area. The Sierra Nevada Range portion of the Mixed Conifer Forest Case Study Area, as well as
19 the Rocky Mountain National Park, have very low amounts of nitrogen deposition (4 kg N/ha/yr
20 for each location), which is consistent with the low amounts of NO_x emissions near these areas.

21 Annual total reactive nitrogen depositions varied by 1 to 3 kg N/ha/yr or less in
22 individual case study areas from 2002 through 2005 (see **Figures 3.3-1a and 3.3-1b**). There is
23 some evidence of a downward trend during this 4-year time period for the six case study areas in
24 the East. No trend is evident for the case study areas in the West. Since the negative effects of
25 nitrogen deposition on sensitive ecosystems may be the result of long-term exposures, recent
26 trends in measured deposition were examined to determine how the amounts of deposition in the
27 2002 analysis year relate to current conditions over a longer time period. As described in Section
28 3-2, trends analyses by CASTNET for an aggregate of 34 sites in the East indicates that dry
29 nitrogen deposition has shown a general decline overall since 2002, but the annual concentration
30 of nitrogen in precipitation has remained fairly steady over this time period (U.S. EPA, 2009). In

1 general, inter-annual variations in meteorology and emissions lead to inter-annual variations in
2 concentrations and deposition.

3 In this section, information available from the NADP National Trends Network¹⁶ on
4 nitrogen deposition for those sites located in and/or near each case study area is examined. To be
5 included in this analysis, the site had to have valid measurements in 2002, as determined by
6 NADP completeness criteria¹⁷. The trend charts for sites selected for this analysis are provided in
7 Appendix 2 of this report. The level of measured annual total wet deposition in 2002 at each site
8 was compared to the amount of deposition in other years over the most recent 10 years (i.e., 1998
9 through 2007)¹⁸. The trend information indicates that overall, for each case study area, the
10 amount of nitrogen deposition in 2002 is generally representative of current conditions.

11 However, deposition trends can vary from site to site, even within a case study area¹⁹.
12 This is most notable for the two sites in the Adirondack Case Study Area and the three sites in
13 the Potomac River/Potomac Estuary Case Study Area. In the Adirondack Case Study Area, the
14 data from the Huntington Wildlife Forest site indicates that wet nitrogen deposition in 2002 is
15 within the range of values measured during other years over the most recent 10-year period. Data
16 from the White Face site shows that wet nitrogen deposition in 2002 was high compared to that
17 in other years. The data at both sites show a downward trend to 2006, with nitrogen deposition
18 increasing again in 2007. For the Potomac River/Potomac Estuary Case Study Area, the trends in
19 wet nitrogen deposition at the Arendtsville, PA, and Parsons, WV, sites indicate that the amount
20 of deposition in 2002 is similar to that from 1998 through 2007. The Wye, MD, site on the
21 Eastern Shore of Maryland shows large inter-annual variations compared to the other sites in the
22 Potomac River/Potomac Estuary Case Study Area and that wet nitrogen deposition in 2002 was
23 on the low end of the range over this time period. In 2002, wet nitrogen deposition for both
24 Sierra Nevada Range portion of the Mixed Conifer Forest Case Study Area, as well as the Rocky
25 Mountain National Park, were within the range of values measured from 1998 through 2007. For
26 the Transverse Range portion of the Mixed Conifer Forest Case Study Area, wet nitrogen
27 deposition was near the low end of the range of values for this period. It is beyond the scope of
28 this analysis to determine the reasons for these differences other than to note that local terrain-

¹⁶ See <http://nadp.sws.uiuc.edu/sites/ntnmap.asp?>

¹⁷ See <http://nadp.sws.uiuc.edu/documentation/completeness.asp>

¹⁸ Some sites do not have historical data back to 1998. For these sites, the amounts of deposition for the available data record were examined.

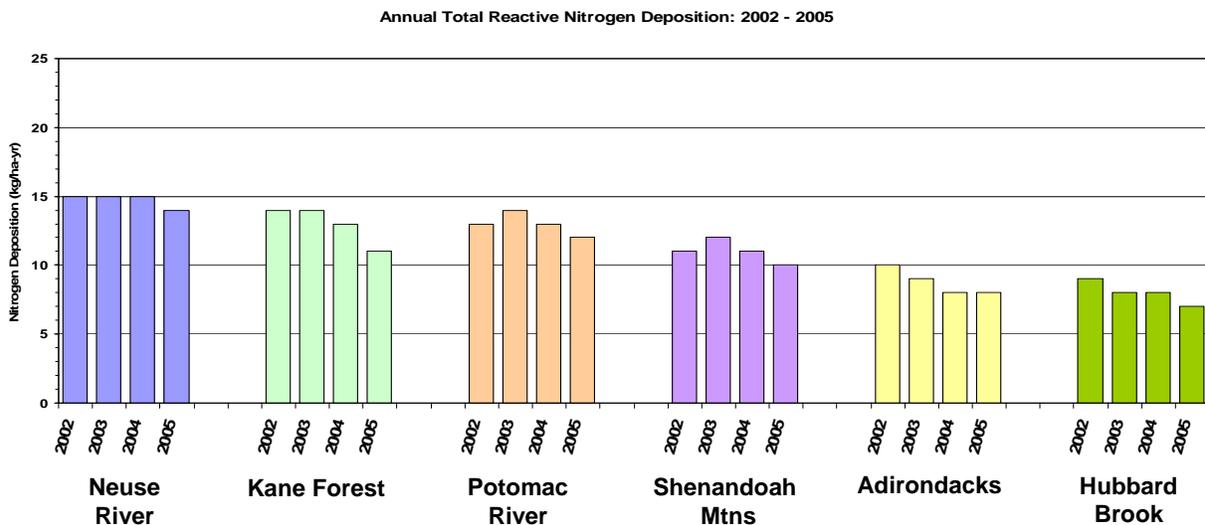
¹⁹ See <http://nadp.sws.uiuc.edu/sites/ntnmap.asp?> for the location of NADP sites across.

- 1 induced meteorological conditions and differential source-receptor relationships across a case
- 2 study area may contribute to the differences noted in deposition trends.

Table 3.3-1. Annual Total Reactive Nitrogen Deposition (kg N/ha/yr) and Sulfur Deposition (kg S/ha/yr) in 2002 for Each Case Study Area, as Well as the Rocky Mountain National Park.

Case Study Areas ^a	2002 Annual Total Deposition	
	Total Reactive Nitrogen (kg N/ha/yr)	Total Sulfur (kg S/ha/yr)
Adirondack	10	9
Hubbard Brook Experimental Forest	8	7
Kane Experimental Forest	13	20
Potomac River/Potomac Estuary	12	14
Shenandoah	11	11
Neuse River/Neuse River Estuary	14	8
Mixed Conifer Forest (Sierra Nevada Range portion)	4	1
Mixed Conifer Forest (Transverse Range portion)	10	2
Rocky Mountain National Park (a supplemental area)	4	1

3 ^a Excludes the Coastal Sage Scrub Case Study Area.



4 **Figure 3.3-1a.** Annual total reactive nitrogen deposition (kg N/ha/yr) from 2002

5 through 2005 for each case study area in the East.

6

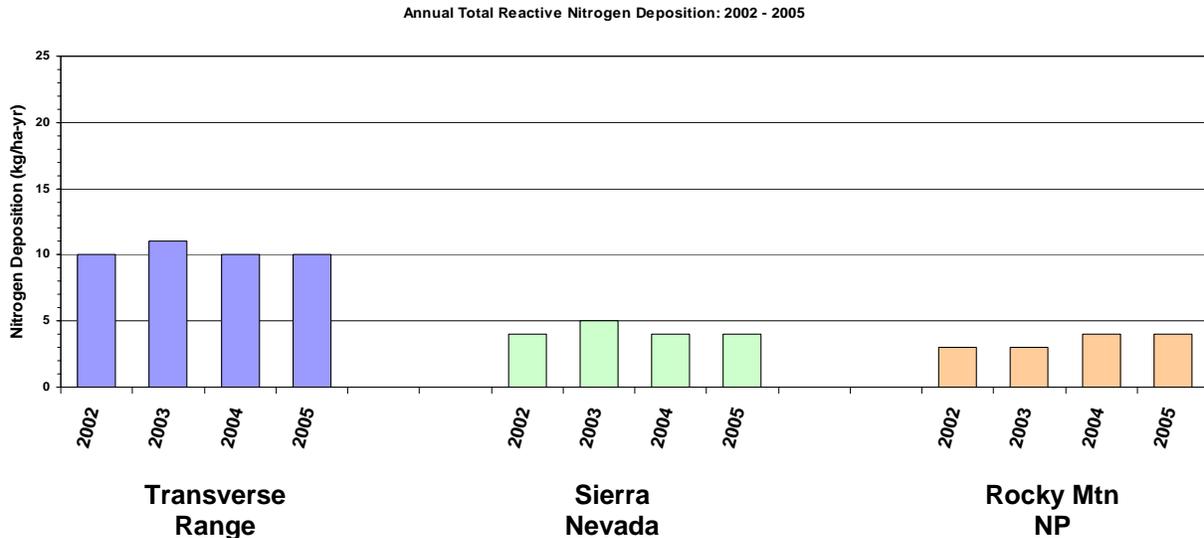


Figure 3.3-1b. Annual total reactive nitrogen deposition (kg N/ha/yr) from 2002 through 2005 for case study areas in the West, and the Rocky Mountain National Park.

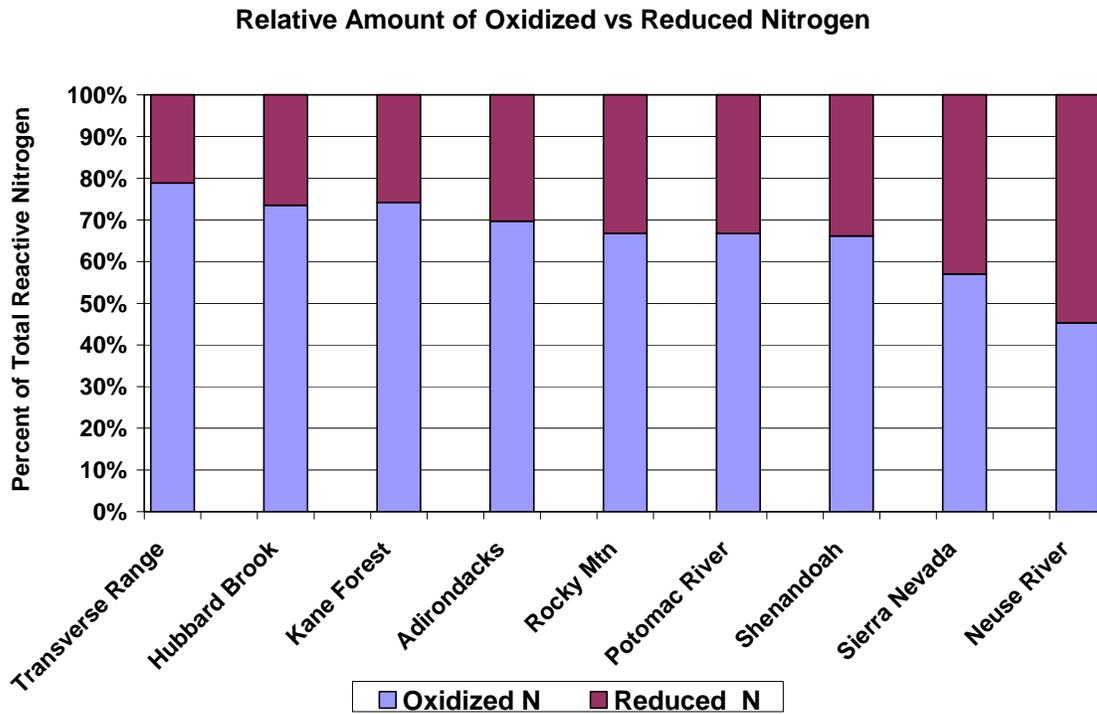
3.3.3.2 Relative Amount of Oxidized and Reduced, Wet and Dry Nitrogen Deposition

The relative amounts of oxidized and reduced nitrogen deposition in 2002 for each case study area, as well as the Rocky Mountain National Park, are shown in **Figure 3.3-2**. Oxidized nitrogen deposition is the dominant contributor to total reactive nitrogen deposition in nearly all of the case study areas. This is consistent with the relative amount of emissions of NO_x and NH_3 . As indicated by comparing **Figures 3.2-2** and **3.2-3**, NO_x emissions are much greater and more widespread compared to emissions of NH_3 , which are more local in nature.

In the Mixed Conifer Forest (Transverse Range portion), Hubbard Brook Experimental Forest, Kane Experimental Forest, and Adirondack case study areas, oxidized nitrogen comprises 70% or more of the total reactive nitrogen. Oxidized nitrogen is 66% to 67% of total reactive nitrogen deposition in the Shenandoah, Potomac River/Potomac Estuary case study areas as well as the Rocky Mountain National Park. In the Neuse River/Neuse River Estuary Case Study Area, reduced nitrogen deposition is >50% of total reactive nitrogen. These findings are consistent with the relative magnitude and geographic distribution of NO_x emissions compared with NH_3 emissions. These findings show that NO_x emissions are much higher than NH_3 emissions in most areas of the country, except near local sources of NH_3 . The relative amount of oxidized versus reduced nitrogen deposition in an area depends on the proximity of the area to local sources of NH_3 . For example, certain portions of the Neuse River/Neuse River Estuary Case Study Area

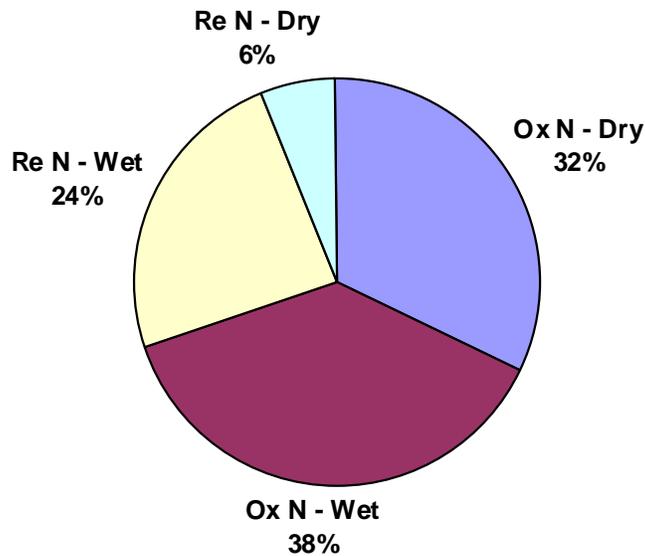
1 contains high NH₃ emissions from hog farm operations, and this area, as a whole, has the largest
2 relative amount of reduced nitrogen deposition. In contrast, the Hubbard Brook Experimental
3 Forest, Kane Experimental Forest, and Adirondack case study areas are distant from sources of
4 high NH₃ emissions, and they each have a low relative amount of reduced nitrogen deposition.

5 The relative amounts of wet and dry, oxidized, and reduced nitrogen for 2002 are shown
6 for each case study area in **Figures 3.3-3(a-i)**. The relative amounts of total reactive nitrogen
7 deposition based on average deposition for the period 2002 through 2005 are shown in Appendix
8 3 of this report. The relative amounts of total reactive nitrogen deposition in 2002 are indicative
9 of conditions over the 4-year period. Looking at the relative amounts of total reactive nitrogen
10 deposition for individual case study areas in the East indicates similar distributions of deposition
11 for several areas. In the Adirondack, Hubbard Brook Experimental Forest, and Kane
12 Experimental Forest case study areas, the relative amount of oxidized nitrogen is about evenly
13 split between wet and dry deposition, whereas the vast majority of reduced nitrogen occurs
14 through wet deposition. In contrast, in the Potomac River/Potomac Estuary and Shenandoah case
15 study areas, dry deposition dominates wet deposition for oxidized nitrogen (~65% of oxidized
16 nitrogen is dry deposited versus 35% wet deposited). However, in these two areas, wet
17 deposition of reduced nitrogen is only slightly greater than dry reduced nitrogen deposition. The
18 Neuse River/Neuse River Estuary Case Study Area is somewhat unique among the case study
19 areas because of the high levels of local NH₃ emissions, which result in a relatively large amount
20 of dry reduced nitrogen deposition compared to the other case study areas in the East. For the
21 two case study areas in the West and the Rocky Mountain National Park, a common feature in
22 the relative amount of nitrogen deposition is that dry oxidized nitrogen is the largest of the four
23 components of total reactive nitrogen deposition at all three of these areas.



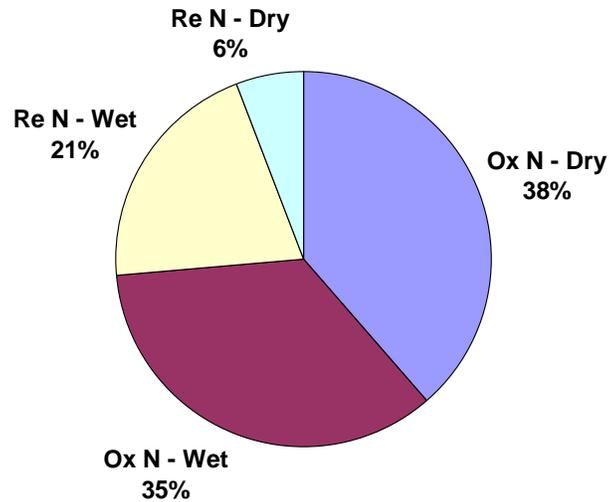
1
 2 **Figure 3.3-2.** Relative amounts of oxidized and reduced nitrogen deposition in
 3 2002 for case study areas and the Rocky Mountain National Park.

Adirondack Case Study Area: 2002 Total Reactive Nitrogen Deposition



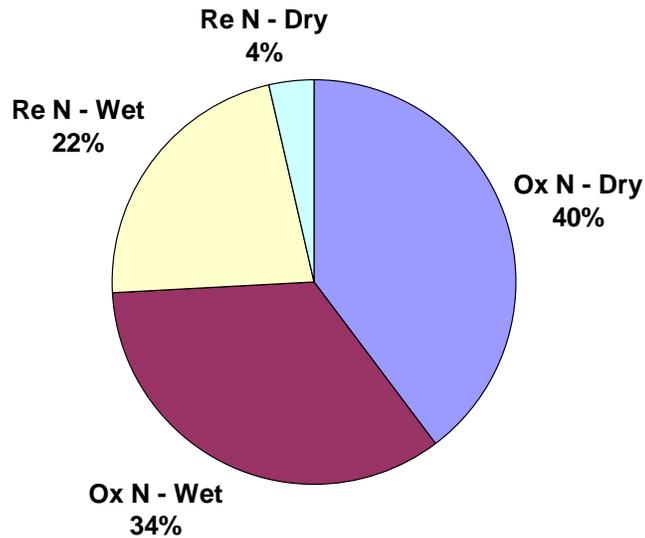
4
 5 **Figure 3.3-3a.** Components of total reactive nitrogen deposition for 2002 in the
 6 Adirondack Case Study Area.

Hubbard Brook Experimental Forest Case Study Area: 2002 Total Reactive Nitrogen Deposition



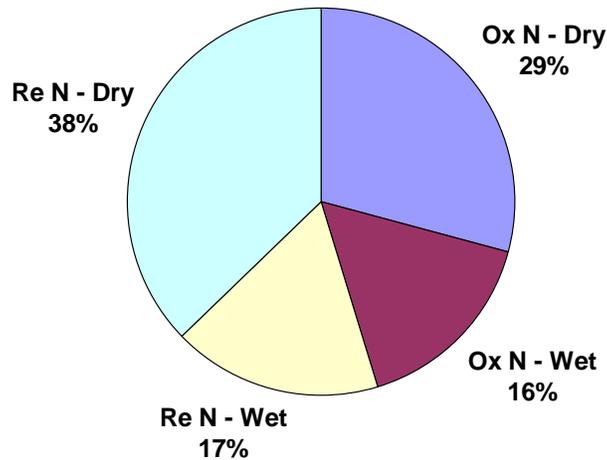
1
2 **Figure 3.3-3b.** Components of total reactive nitrogen deposition for 2002 in the
3 Hubbard Brook Experimental Forest Case Study Area.

Kane Experimental Forest Case Study Area: 2002 Total Reactive Nitrogen Deposition



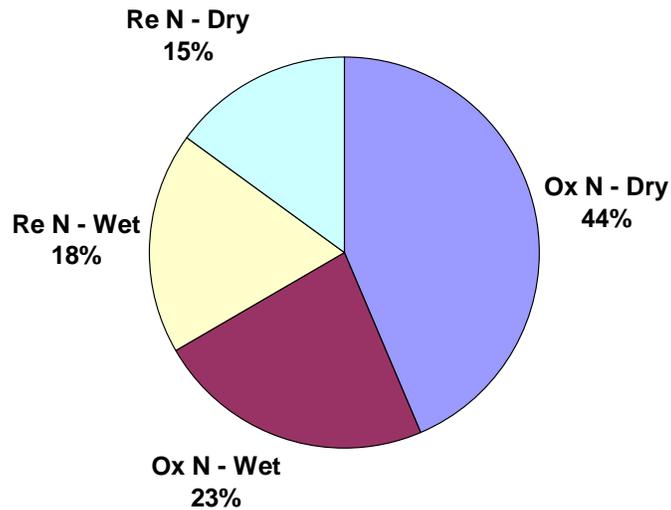
4
5 **Figure 3.3-3c.** Components of total reactive nitrogen deposition for 2002 in the
6 Kane Experimental Forest Case Study Area.

Neuse River/Neuse River Estuary Case Study Area: 2002 Total Reactive Nitrogen Deposition



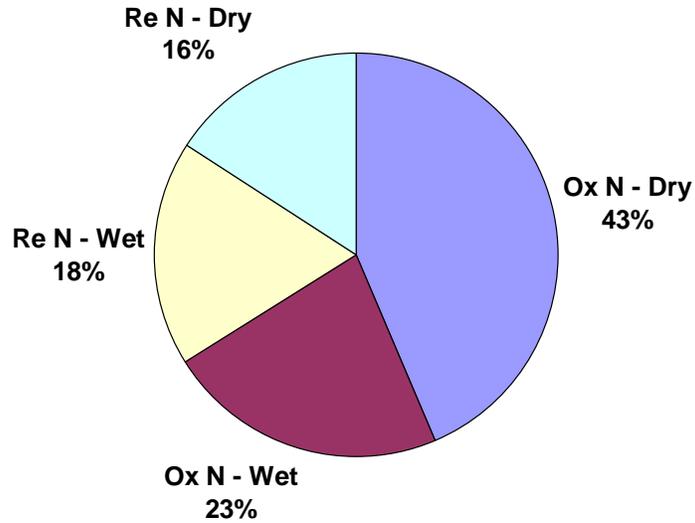
1
2 **Figure 3.3-3d.** Components of total reactive nitrogen deposition for 2002 in the
3 Neuse River/Neuse River Estuary Case Study Area.

Potomac River/Potomac Estuary Case Study Area: 2002 Total Reactive Nitrogen Deposition



4
5 **Figure 3.3-3e.** Components of total reactive nitrogen deposition for 2002 in the
6 Potomac River/Potomac Estuary Case Study Area.

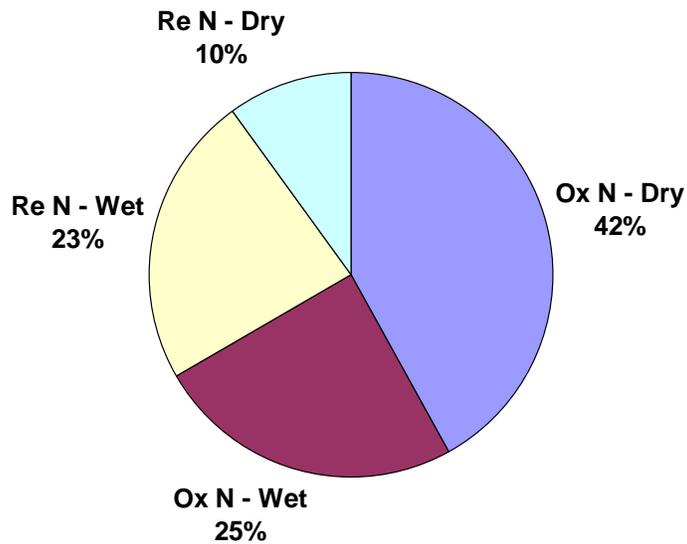
Shenandoah Case Study Area: 2002 Total Reactive Nitrogen Deposition



1
2
3

Figure 3.3-3f. Components of total reactive nitrogen deposition for 2002 in the Shenandoah Case Study Area.

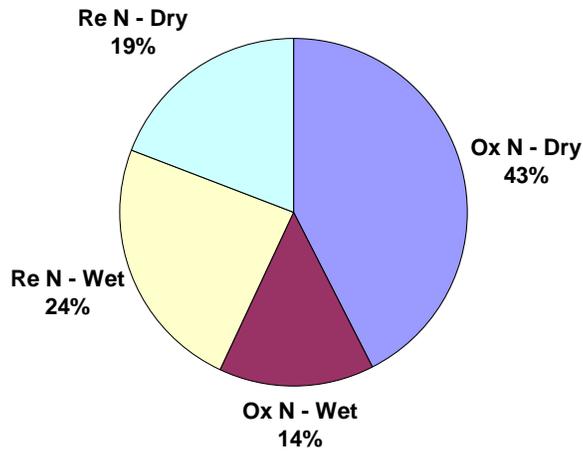
Rocky Mountain National Park: 2002 Total Reactive Nitrogen Deposition



4
5
6

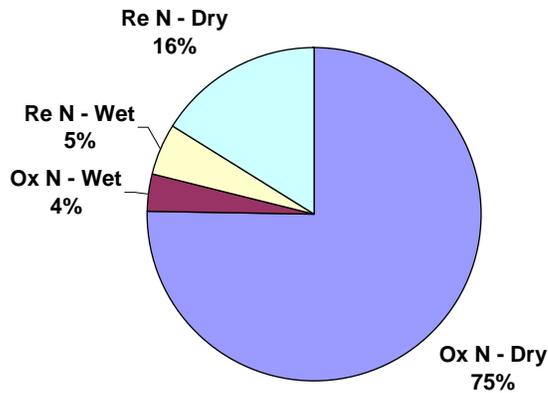
Figure 3.3-3g. Components of total reactive nitrogen deposition for 2002 in the Rocky Mountain National Park.

Mixed Conifer Forest (Sierra Nevada Range) Case Study Area: 2002 Total Reactive Nitrogen Deposition



1
2 **Figure 3.3-3h.** Components of total reactive nitrogen deposition for 2002 in the
3 Sierra Nevada Range portion of the Mixed Conifer Forest Case Study Area.

Mixed Conifer Forest (Transverse Range) Case Study Area: 2002 Total Reactive Nitrogen Deposition



4
5 **Figure 3.3-3i.** Components of total reactive nitrogen deposition for 2002 in the
6 Transverse Range portion of the Mixed Conifer Forest Case Study Area.

1 **3.3.3.3 Geographic Variations in Annual Total Reactive Nitrogen Deposition for**
2 **2002²⁰**

3 The geographic variations in total reactive, oxidized, reduced, wet, and dry nitrogen
4 deposition in 2002 are shown in **Figures 3.3-4a-e** for the case study areas in the East²¹.

5 **Figures 3.3-4a-c** shows the geographic variations in total reactive nitrogen deposition and
6 oxidized and reduced nitrogen deposition for the West²².

7 **Adirondack Case Study Area**

8 As shown in **Figure 3.3-4a**, total reactive nitrogen deposition in 2002 decreases from
9 southwest to northeast across the Adirondack Case Study Area. For example, total reactive
10 nitrogen deposition is >12 kg N/ha/yr in the southwestern portion of the Adirondack Case Study
11 Area compared to <8 kg N/ha/yr in some parts of the eastern portion of this area. By comparing
12 the oxidized nitrogen deposition map in **Figure 3.3-4b** to the reduced nitrogen deposition map in
13 **Figure 3.3-4c**, it is evident that oxidized nitrogen deposition is much greater than reduced
14 nitrogen across the entire case study area. Oxidized nitrogen values are generally in the range of
15 5 to 7 kg N/ha/yr, with values of 7 to 9 kg N/ha/yr in the southwestern part of the area. In
16 contrast, reduced nitrogen deposition is generally 2 to 3 kg N/ha/yr. From **Figure 3.3-4a**, it is
17 evident that the relatively high total reactive nitrogen deposition in the far southwestern portion
18 of this case study area is part of a broad area of high nitrogen deposition that stretches westward
19 from the Adirondack Case Study Area along the southern shore of Lake Ontario toward western
20 Pennsylvania and Ohio.

21 The spatial patterns in wet and dry nitrogen are shown in **Figure 3.3-4d** and
22 **Figure 3.3-4e**, respectively. Wet deposition is in the range of 5 to 7 kg N/ha/yr across the region,
23 with higher amounts in the southwestern section. Dry deposition is lower than wet deposition
24 overall and declines fairly rapidly from values of 4 to 5 kg N/ha/yr in the western portion to 2 to
25 3 kg N/ha/yr in the eastern portion.

²⁰ Note that an analysis of the spatial gradients in reactive nitrogen and sulfur deposition for the Kane Experimental Forest and Hubbard Brook Experimental Forest case study areas, as well as the Rocky Mountain National Park is not included because the size of each of these areas is small relative to the 12 x 12-km resolution-measured data and model predictions used in this analysis.

²¹ Deposition in all of the figures is displayed at a resolution of 12 x 12 km to be consistent with the aggregated wet and dry deposition data sets described above.

²² Because of the highly rugged terrain in the case study areas of the West, there is less confidence that the 12-km data represents the true geographic variations in deposition. This is particularly true for wet deposition, which is based on spatial interpolation from a relatively sparse monitoring network. Thus, a discussion of the geographic variations in wet and dry deposition for the case study areas in the West is not included.

1 **Shenandoah Case Study Area**

2 As shown in **Figure 3.3-4a**, total reactive nitrogen deposition in the southern portion of
3 the Shenandoah Case Study Area is in the range of 8 to 10 kg N/ha/yr, increasing to ≥ 14 kg
4 N/ha/yr for the northern portions. Oxidized nitrogen ranges from 5 to 9 kg N/ha/yr, which is
5 greater than the reduced nitrogen deposition in most of this area. However, the highest levels of
6 nitrogen deposition found in the northern portion are mostly due to reduced nitrogen deposition,
7 which can be seen by comparing **Figure 3.3-4b** with **Figure 3.3-4c**. The higher reduced nitrogen
8 deposition (>9 kg N/ha/yr) is largely the result of high NH₃ emissions in this northern portion of
9 this case study area, as shown in **Figure 3.2-3**. These NH₃ emissions are associated with poultry
10 farm operations in this general location. Elsewhere across the Shenandoah Case Study Area,
11 reduced nitrogen deposition is in the range of 2 to 3 kg N/ha/yr.

12 Over most of the Shenandoah Case Study Area, wet nitrogen deposition in 2002 is in the
13 range of 4 to 5 kg N/ha/yr, with lower amounts of 3 to 4 kg N/ha/yr in parts of the southern
14 portion of this area. In contrast, dry nitrogen deposition exhibits a peak of relatively high NH₃
15 emissions in the northern portion of the area. There, the amount of dry nitrogen deposition is 14
16 kg N/ha/yr or greater.

17 **Potomac River/Potomac Estuary Case Study Area**

18 As shown in **Figure 3.3-4a**, there are large spatial variations in annual total reactive
19 nitrogen deposition across the Potomac River/Potomac Estuary Case Study Area. The highest
20 levels of total reactive nitrogen deposition in 2002 are seen in the portion of this area over
21 northwestern Virginia and from southern Pennsylvania to the Baltimore-Washington, DC,
22 metropolitan area. In these portions of this case study area, annual total reactive nitrogen
23 deposition exceeds 14 kg N/ha/yr. Between these areas of high deposition, total reactive nitrogen
24 deposition declines to the general range of 10 to 12 kg N/ha/yr.

25 The spatial patterns in oxidized and reduced nitrogen deposition are shown in
26 **Figures 3.3-4b** and **3.3-4c**. From these figures, it is clear that oxidized nitrogen deposition is
27 greater than reduced nitrogen deposition across most of this case study area. Oxidized nitrogen
28 deposition is in the range of 9 to 14 kg N/ha/yr in and near the Baltimore-Washington, DC, urban
29 area. Oxidized nitrogen levels decline from east to west across the remainder of this case study
30 area down to the range of 5 to 7 kg N/ha/yr over the western portions of this area. The localized
31 high levels of reduced nitrogen deposition correspond to the locations of high NH₃ emissions, as

1 shown in **Figure 3.2-3**. Elsewhere in this case study area, reduced nitrogen deposition is fairly
2 low, mostly in the range of 3 to 4 kg N/ha/yr.

3 The patterns of wet nitrogen deposition in the Potomac River/Potomac Estuary Case
4 Study Area indicate that in 2002, the northern portion of this area had higher amounts of wet
5 nitrogen deposition (5 to 7 kg N/ha/yr) compared with the southern portion (4 to 5 kg N/ha/yr).
6 Dry deposition was highest in the vicinity of the high NH₃ emissions in the far southwestern
7 portion of this area. Relatively large amounts of dry nitrogen deposition are also seen in the
8 eastern half of this area. Considering the spatial distribution of NO_x and NH₃ emissions in and
9 near the Potomac River, it appears that NH₃ emissions from livestock farms in south-central
10 Pennsylvania may be contributing to the higher amounts of dry nitrogen deposition close to the
11 Maryland-Pennsylvania border. In contrast, the high NO_x emissions near the Washington, DC,
12 area may be contributing to the relatively high dry nitrogen deposition in this part of the Potomac
13 River/Potomac Estuary Case Study Area.

14 **Neuse River/Neuse River Estuary Case Study Area**

15 The central portions of the Neuse River/Neuse River Estuary Case Study Area are
16 impacted by high amounts of total reactive nitrogen deposition in amounts >20 kg N/ha/yr (see
17 **Figure 3.3-4a**). These high levels of deposition are associated with high NH₃ emissions from
18 swine and poultry production facilities in the southeastern part of North Carolina (see **Figure**
19 **3.2-3**). In contrast to the large spatial gradients seen in reduced nitrogen deposition, oxidized
20 nitrogen deposition is fairly homogenous across this case study area. Most of the area has
21 oxidized nitrogen deposition in the range of 5 to 7 kg N/ha/yr, which increases to 7 to 9 kg
22 N/ha/yr near the Raleigh-Durham urban area.

23 Wet and dry nitrogen deposition in the Neuse River/Neuse River Estuary Case Study
24 Area show similar patterns in that the highest amounts of deposition are in the vicinity of high
25 NH₃ emissions near the central portion of this area. The lowest amounts of wet and dry nitrogen
26 deposition are near the coast.

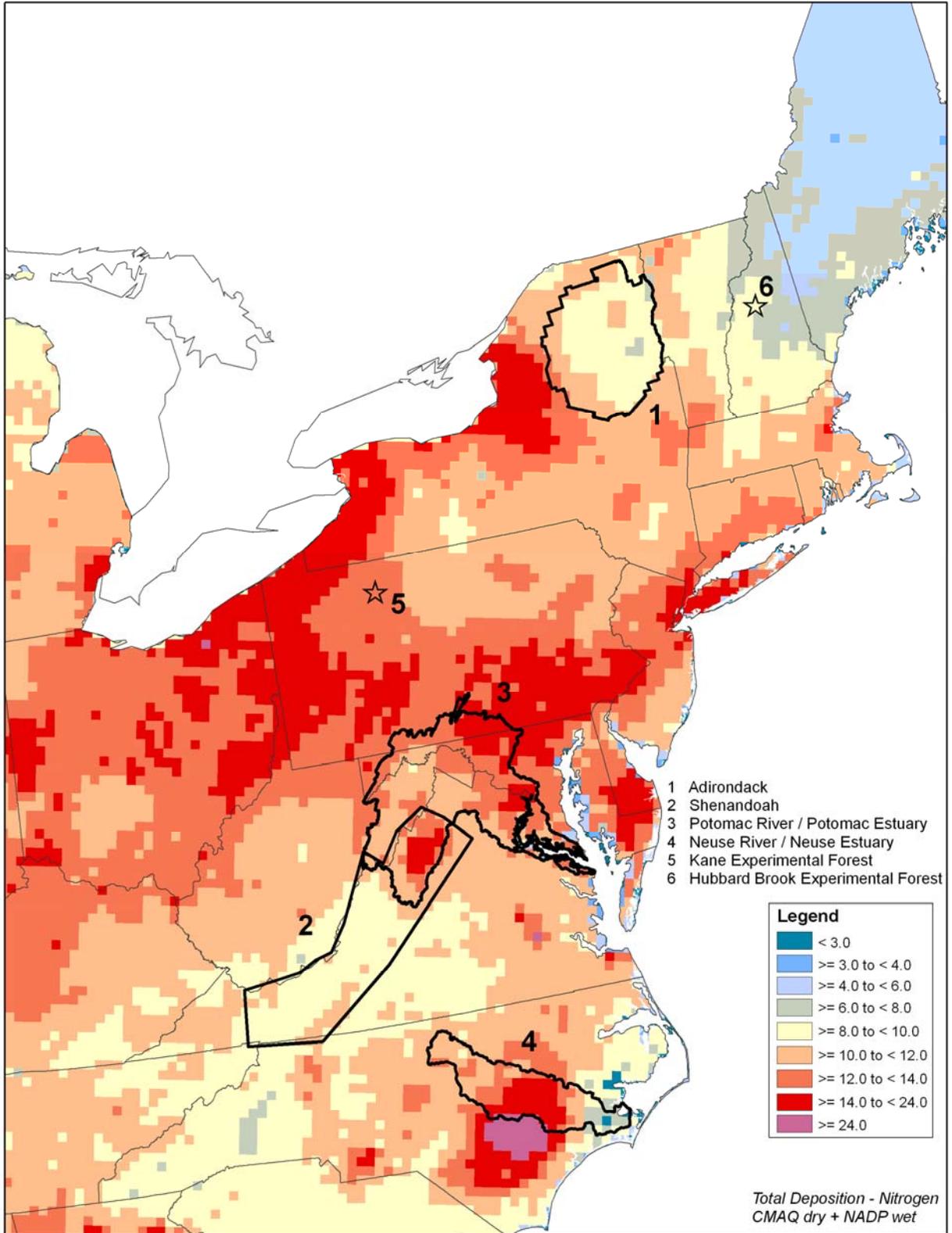
27 **Sierra Nevada Range (a Portion of the Mixed Conifer Forest Case Study Area)**

28 As seen from **Figure 3.3-5a**, there is a west to east gradient in total reactive nitrogen
29 deposition across the Sierra Nevada Range portion of the Mixed Conifer Forest Case Study Area.
30 In the extreme western portion of this area, which is near the San Joaquin Valley, total reactive
31 nitrogen depositions are in the range of 6 to 8 kg N/ha/yr. Total reactive nitrogen deposition
32 declines to the range of 2 to 3 kg N/ha/yr in the eastern half of this case study area. Both

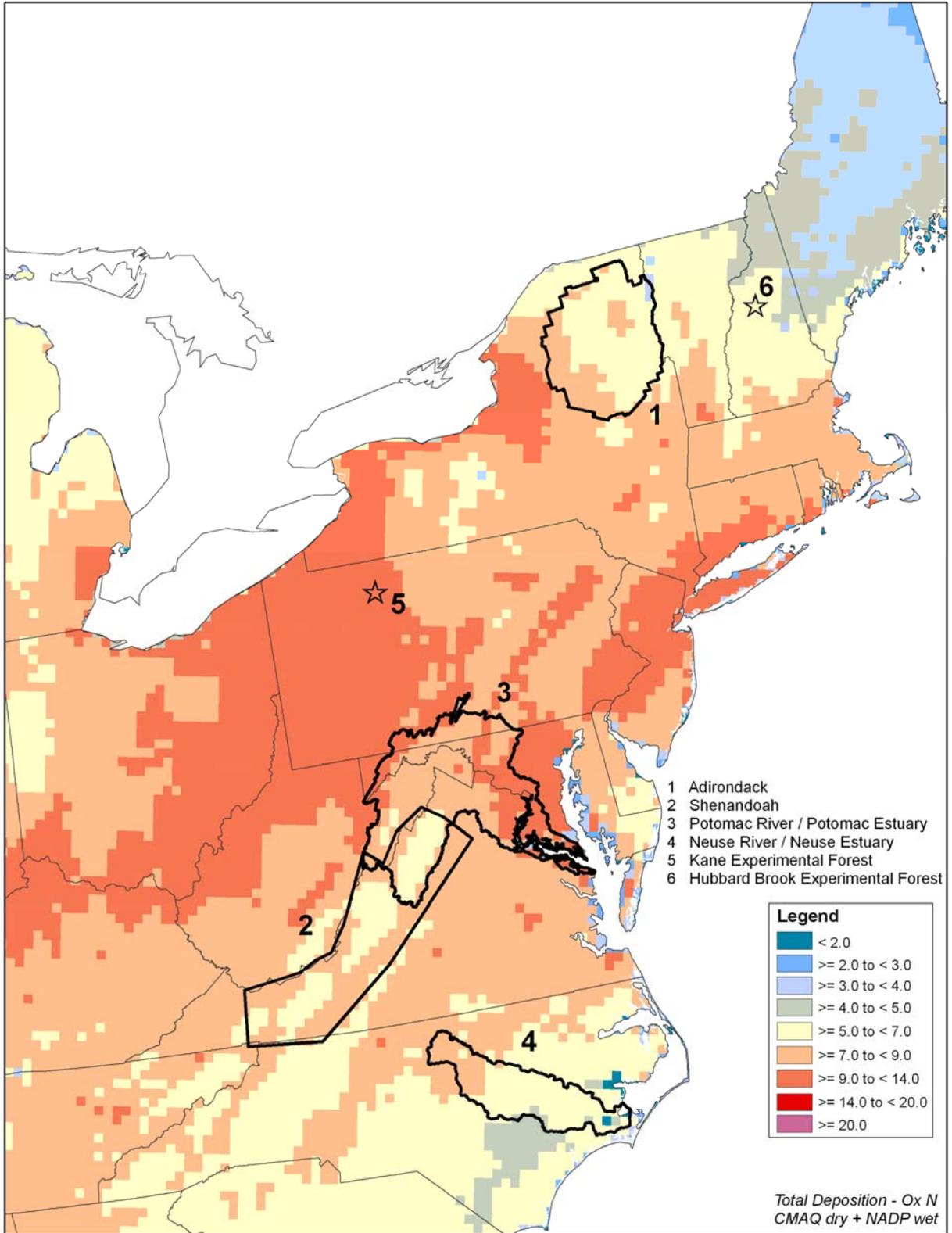
1 oxidized and reduced nitrogen deposition exhibit similar west to east gradient in deposition as
2 seen in **Figures 3.3-5b** and **3.3-5c**.

3 **Transverse Range (a Portion of the Mixed Conifer Forest Case Study Area)**

4 High amounts of total reactive nitrogen deposition are evident across much of the
5 Transverse Range portion of the Mixed Conifer Forest Case Study Area as evident in **Figure 3.3-**
6 **5a**. This figure shows total reactive nitrogen deposition levels of 12 kg N/ha/yr or greater over
7 portions of the San Bernardino Mountains to the west and northwest of the Los Angeles urban
8 area. As indicated above, oxidized nitrogen deposition is much greater than reduced nitrogen
9 deposition throughout nearly all of this case study area. The large amounts of oxidized nitrogen
10 deposition are associated with the high levels of NO_x emissions in this portion of southern
11 California, as seen in **Figure 3.2-2**.

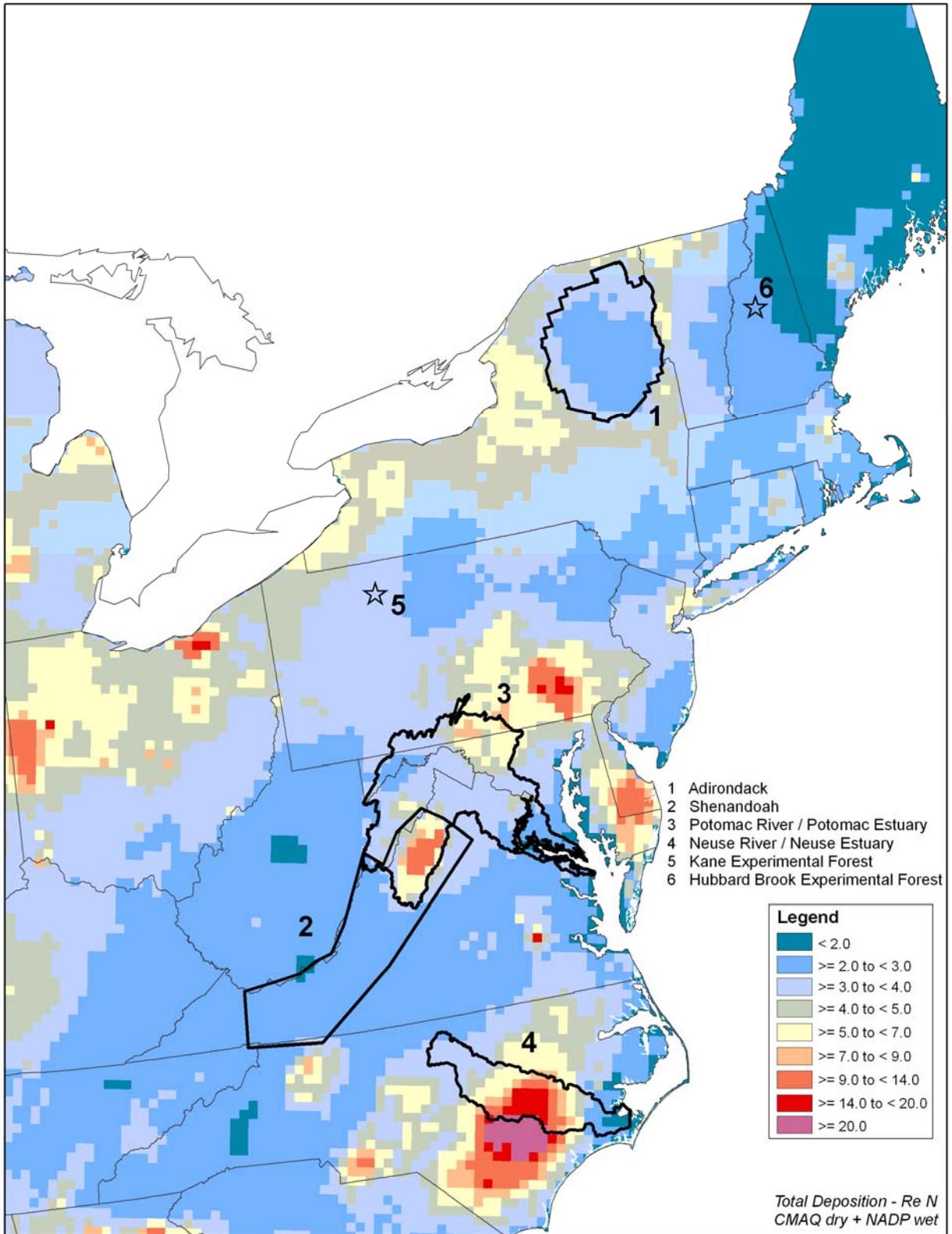


1
 2 **Figure 3.3-4a.** Annual total dry plus wet reactive nitrogen deposition (kg N/ha/yr)
 3 in 2002 for the case study areas in the East.

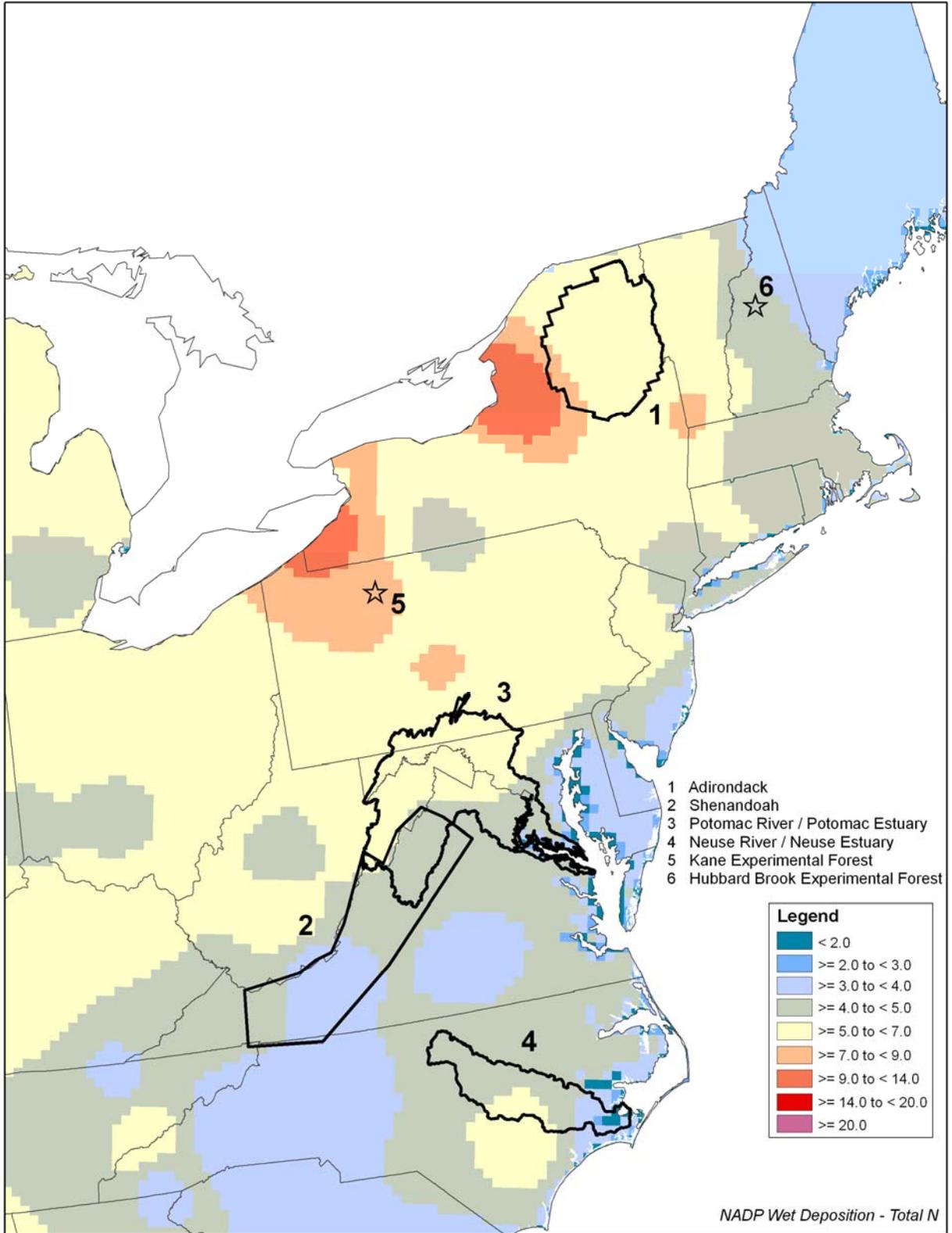


1
 2
 3

Figure 3.3-4b. Annual total dry plus wet oxidized nitrogen deposition (kg N/ha/yr) in 2002 for the case study areas in the East.

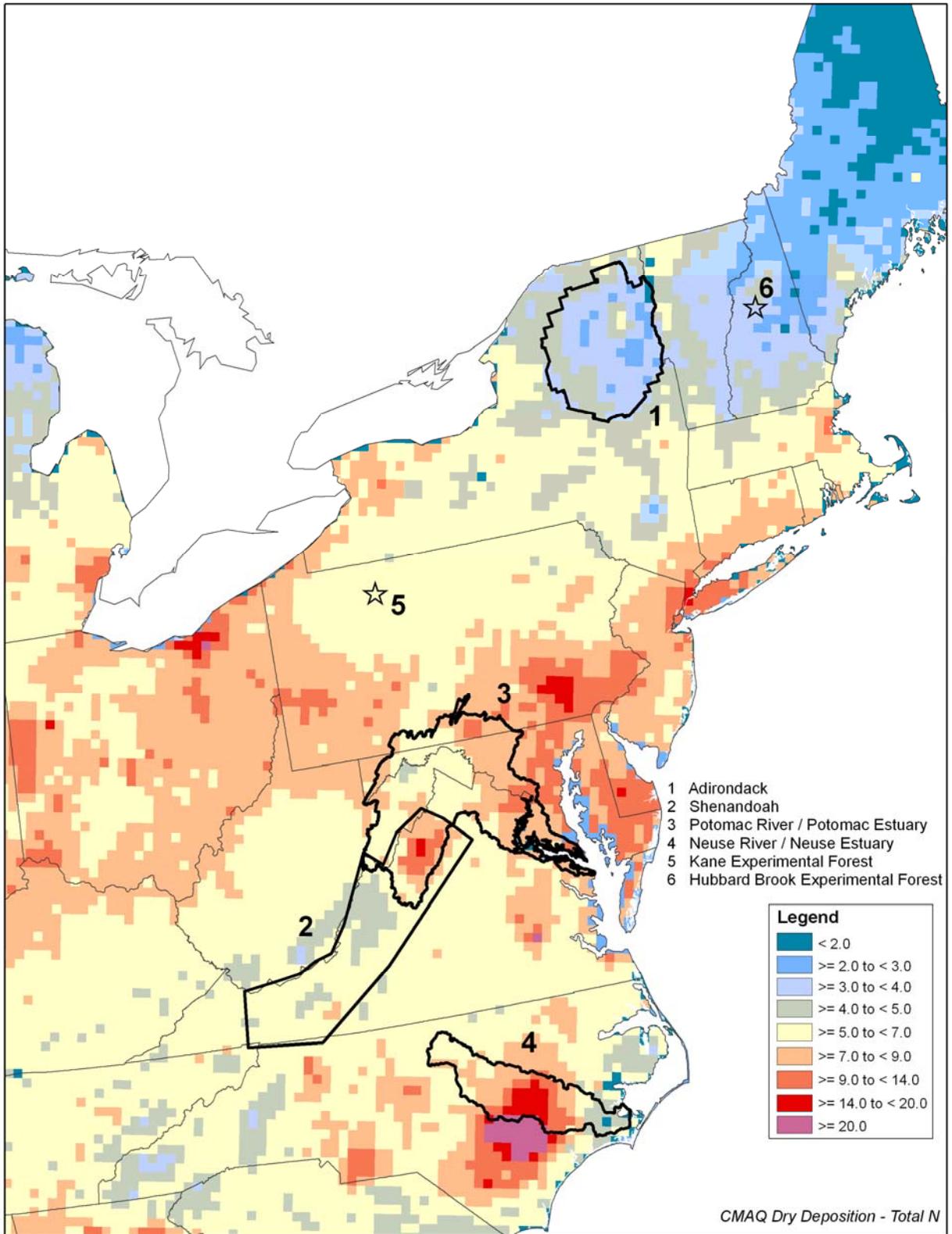


1
 2 **Figure 3.3-4c.** Annual total dry plus wet reduced nitrogen deposition (kg N/ha/yr)
 3 in 2002 for the case study areas in the East.

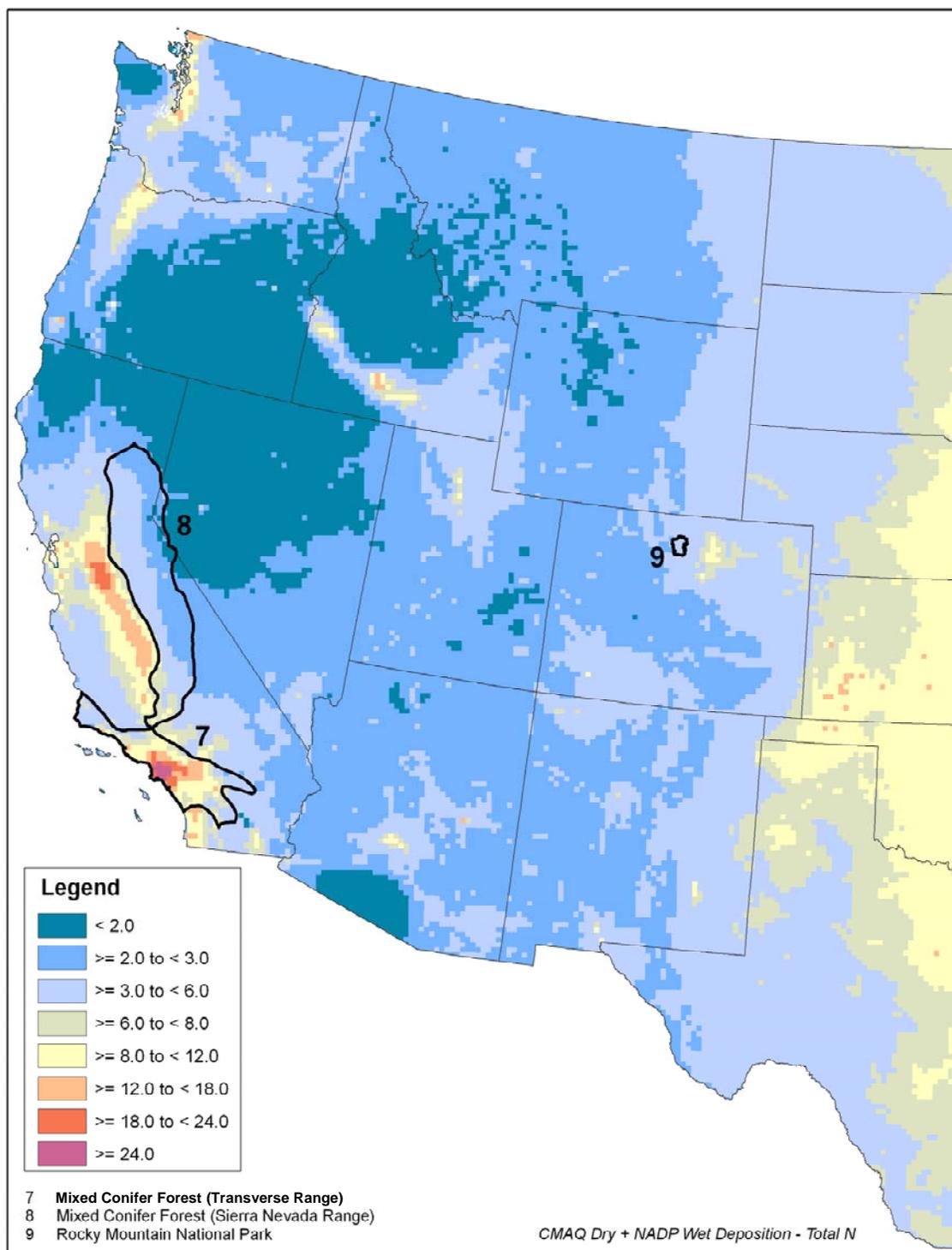


1
 2
 3

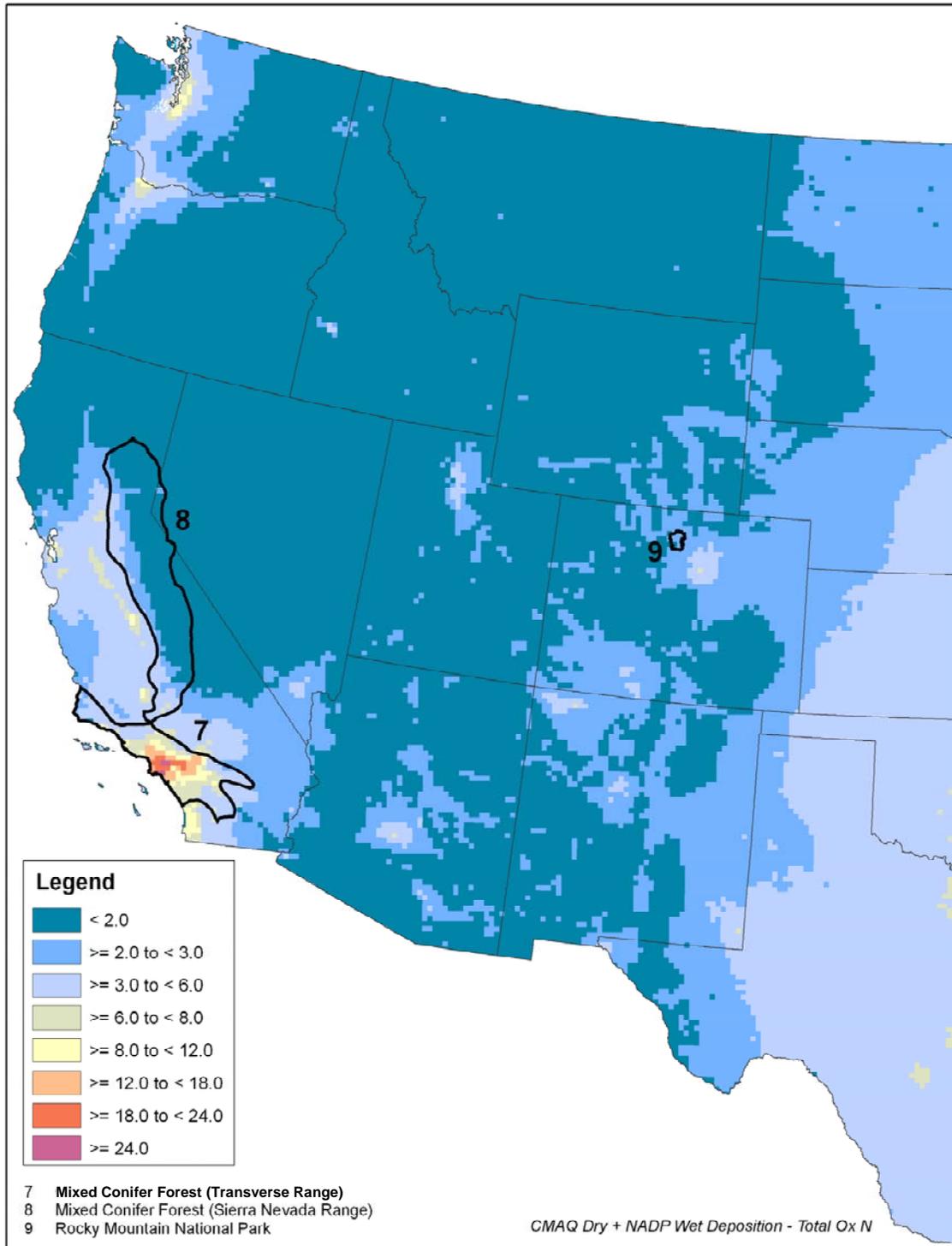
Figure 3.3-4d. Annual total wet reactive nitrogen deposition (kg N/ha/yr) in 2002 for the case study areas in the East.



1
 2 **Figure 3.3-4e.** Annual total dry reactive nitrogen deposition (kg N/ha/yr) in 2002
 3 for the case study areas in the East.

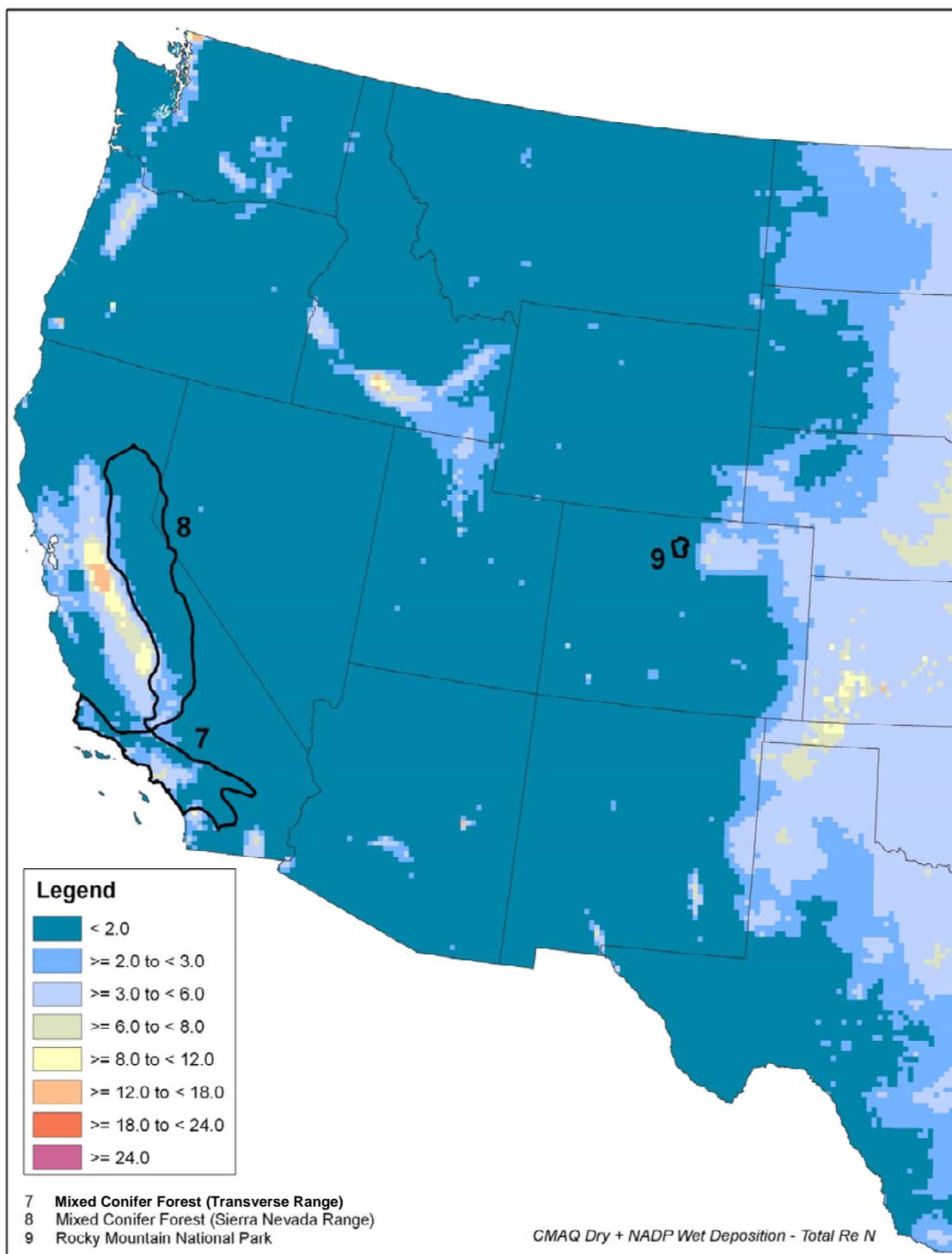


1
 2 **Figure 3.3-5a.** Annual total dry plus wet reactive nitrogen deposition (kg N/ha/yr)
 3 in 2002 for case study areas and Rocky Mountain National Park in the West.



1
2
3
4

Figure 3.3-5b. Annual total dry plus wet oxidized nitrogen deposition (kg N/ha/yr) in 2002 for case study areas and Rocky Mountain National Park in the West.



1
 2 **Figure 3.3-5c.** Annual total dry plus wet reduced nitrogen deposition (kg N/ha/yr)
 3 in 2002 for case study areas and Rocky Mountain National Park in the West.

1 **3.3.3.4 Seasonal Variations in Total Reactive Nitrogen Deposition for 2002**

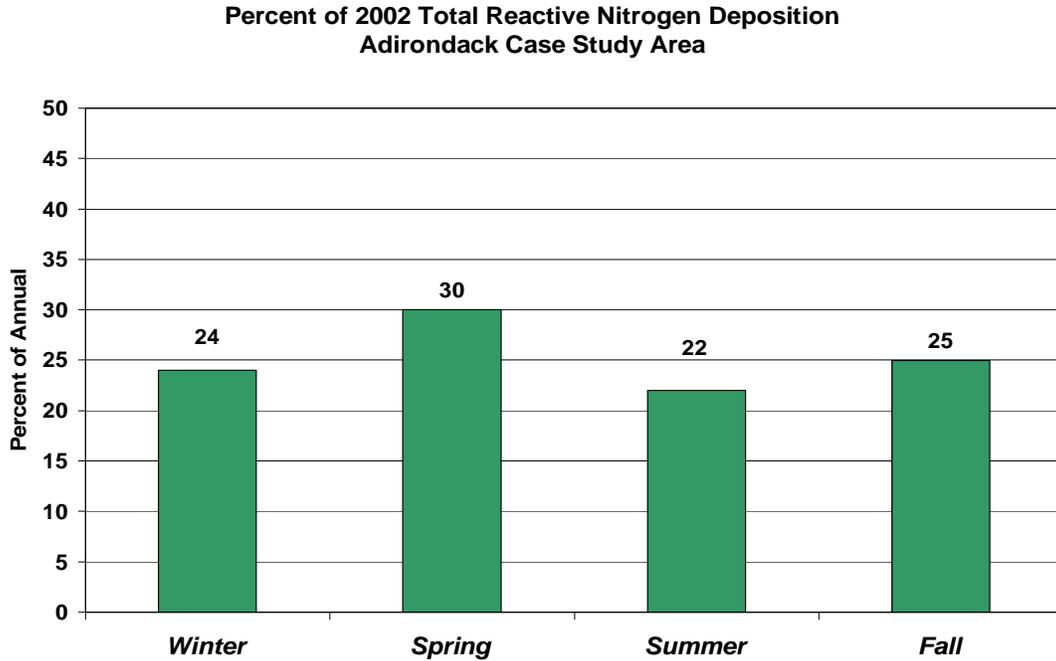
2 The seasonal variations in model-predicted 2002 total reactive nitrogen deposition for
3 each case study area are shown in **Figures 3.3-6(a-i)**. In most of the case study areas, total
4 reactive nitrogen is highest in spring or summer. Among the case study areas in the East, total
5 reactive nitrogen is highest in spring for the Adirondack, Hubbard Brook Experimental Forest,
6 and Kane Experimental Forest case study areas. In these areas, total reactive nitrogen deposition
7 in spring is 30% or more of the annual total. The temporal variation in total reactive nitrogen
8 deposition is fairly flat in the other three seasons (20% to 25% of the annual total). The results on
9 the seasonal patterns in nitrogen deposition for the case study areas in the East are generally with
10 the findings by Sickles and Shadwick (2007b). In the West, the seasonal variations in the Sierra
11 Nevada Range portion of the Mixed Conifer Forest Case Study Area and the Rocky Mountain
12 National Park are similar, with a peak in spring and relatively high amounts of deposition also
13 seen in summer. Total reactive nitrogen deposition is highest in summer in the Transverse Range
14 portion of the Mixed Conifer Forest Case Study Area.

15 The seasonal variations in total reactive nitrogen deposition reflect the aggregate of the
16 variations in dry and wet, oxidized, and reduced nitrogen deposition, which are shown in
17 **Figures 3.3-7(a-i)**²³. Seasonal patterns in precipitation²⁴ for each case study area are also shown
18 in **Figures 3.3-7(a-i)**. Dry oxidized nitrogen deposition peaks in spring or summer and tends to
19 have the least seasonal variation among the four components of total reactive nitrogen
20 deposition. In contrast, reduced nitrogen deposition peaks in summer and exhibits a fairly large
21 seasonal variation in each of the case study areas. The amount of reduced nitrogen dry deposition
22 in summer accounts for >40% of the annual total reduced nitrogen dry deposition in each area,
23 except for the Kane Experimental Forest Case Study Area and the Transverse Range portion of
24 the Mixed Conifer Forest Case Study Area, where in summer, dry reduced nitrogen is 30% to
25 35% of the annual total. The intra-annual variations in dry reduced nitrogen deposition are
26 generally consistent with the temporal patterns in NH₃ emissions, which exhibit a primary peak
27 in summer and a secondary peak in spring for the states in which the case study areas are located,
28 as shown in **Figure 3.3-8**. Wet reduced nitrogen deposition seasonal variations generally, but not
29 always, align with the seasonal variations in precipitation. Seasonal variations in wet oxidized

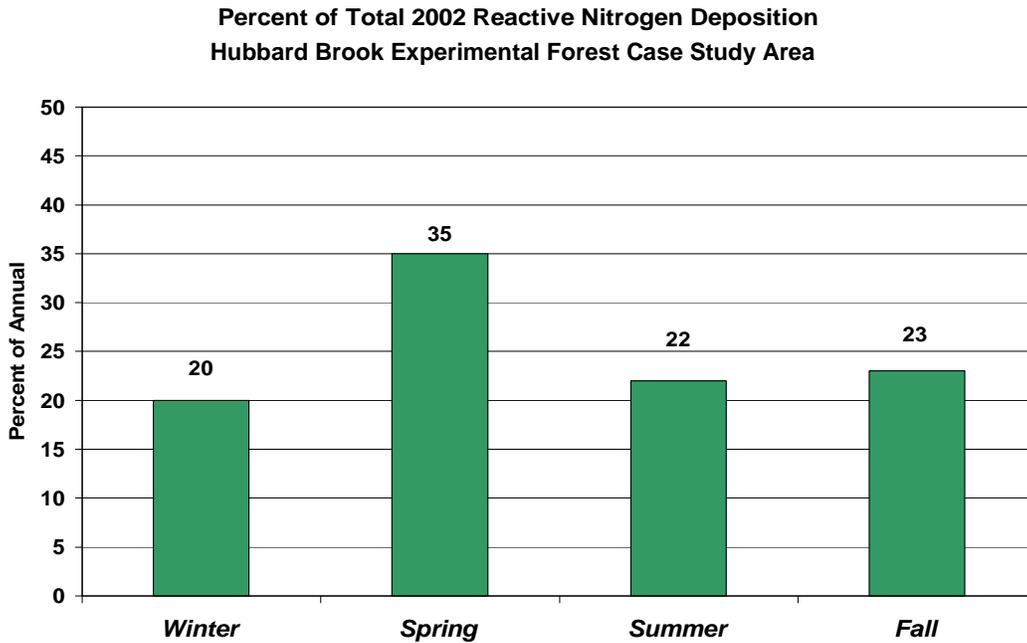
²³ In these figures the percent of deposition by season for each category sums to 100 percent.

²⁴ The precipitation data used in this analysis are based on the MM-5 meteorological model predictions, which are used as inputs to the CMAQ model simulations.

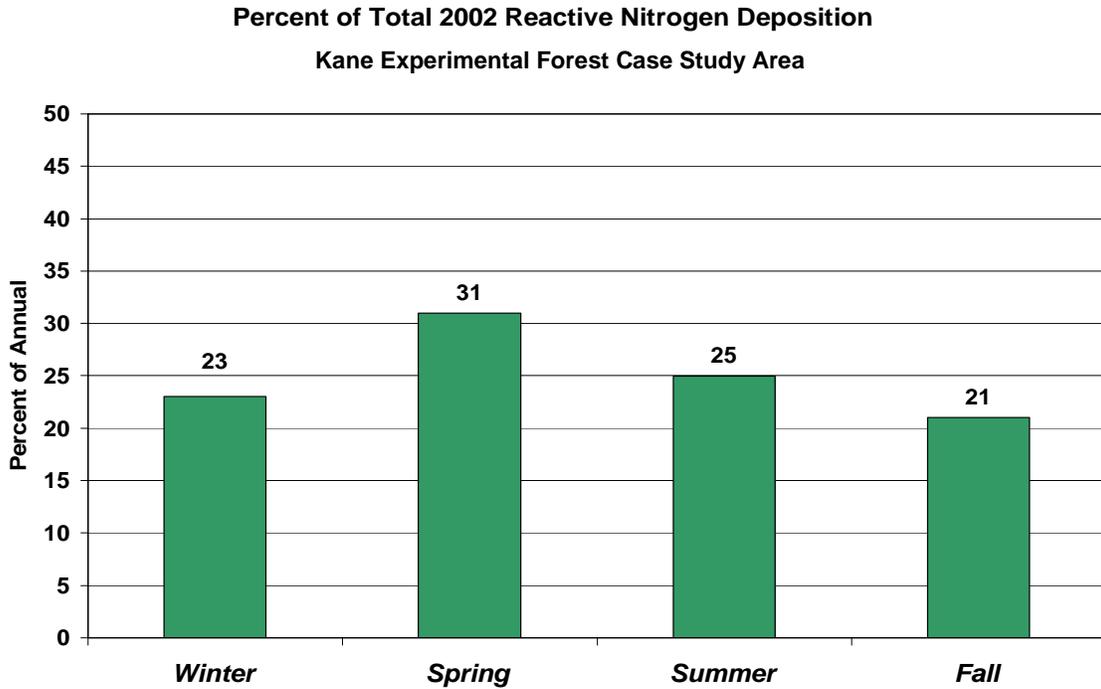
- 1 nitrogen deposition also appear to reflect precipitation patterns, but not as closely as do wet
- 2 reduced nitrogen deposition.



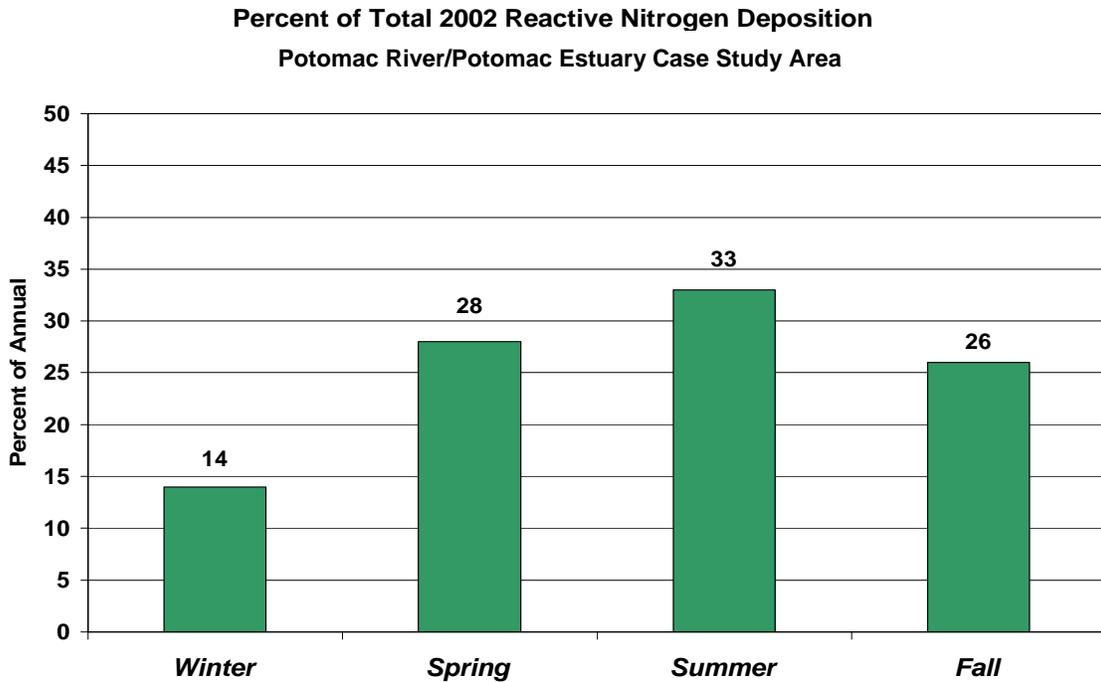
3
4 **Figure 3.3-6a.** Percentage of 2002 total reactive nitrogen deposition in the
5 Adirondack Case Study Area.



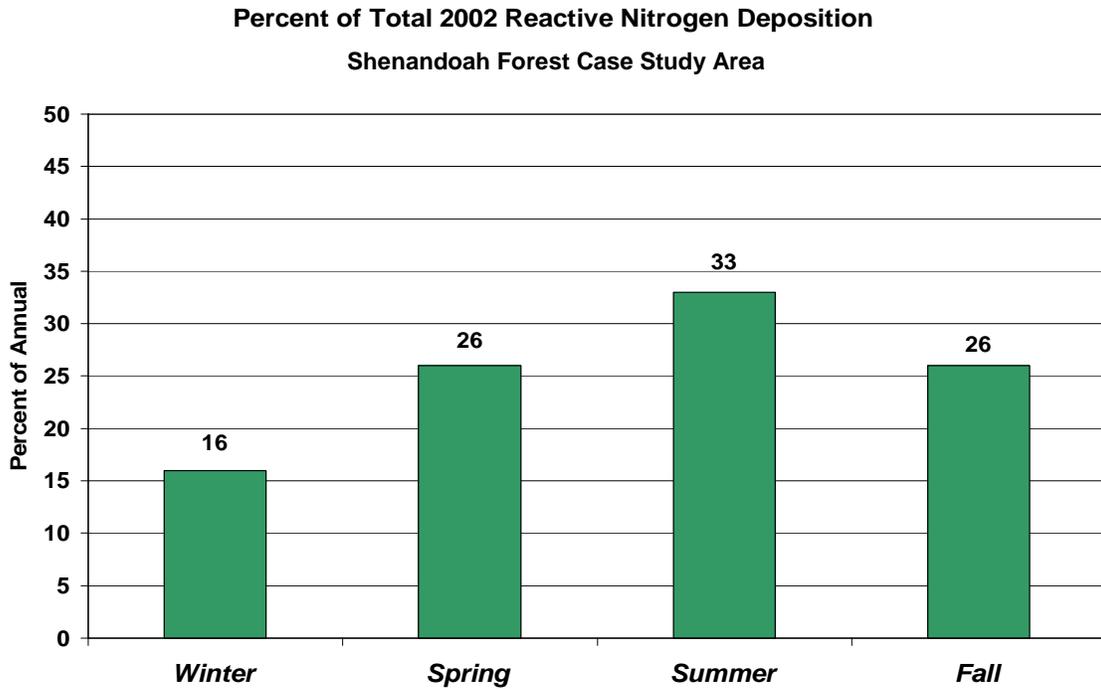
6
7 **Figure 3.3-6b.** Percentage of 2002 total reactive nitrogen deposition in the
8 Hubbard Brook Experimental Forest Case Study Area.



1
2 **Figure 3.3-6c.** Percentage of 2002 total reactive nitrogen deposition in the Kane
3 Experimental Forest Case Study Area.

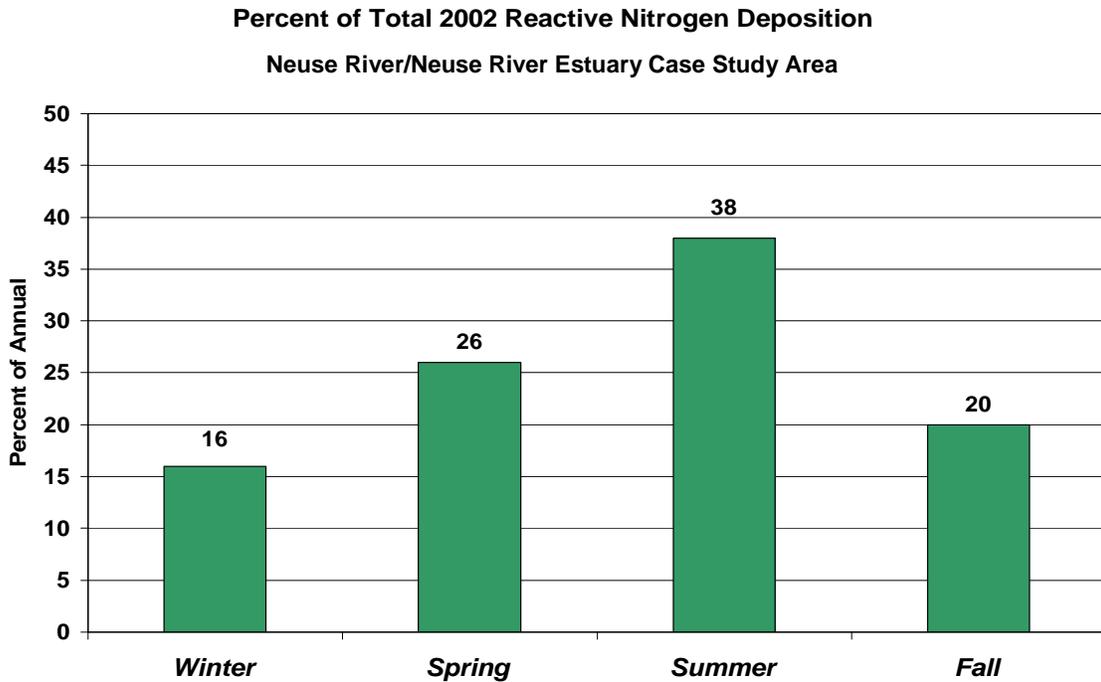


4
5 **Figure 3.3-6d.** Percentage of 2002 total reactive nitrogen deposition in the
6 Potomac River/Potomac Estuary Case Study Area.



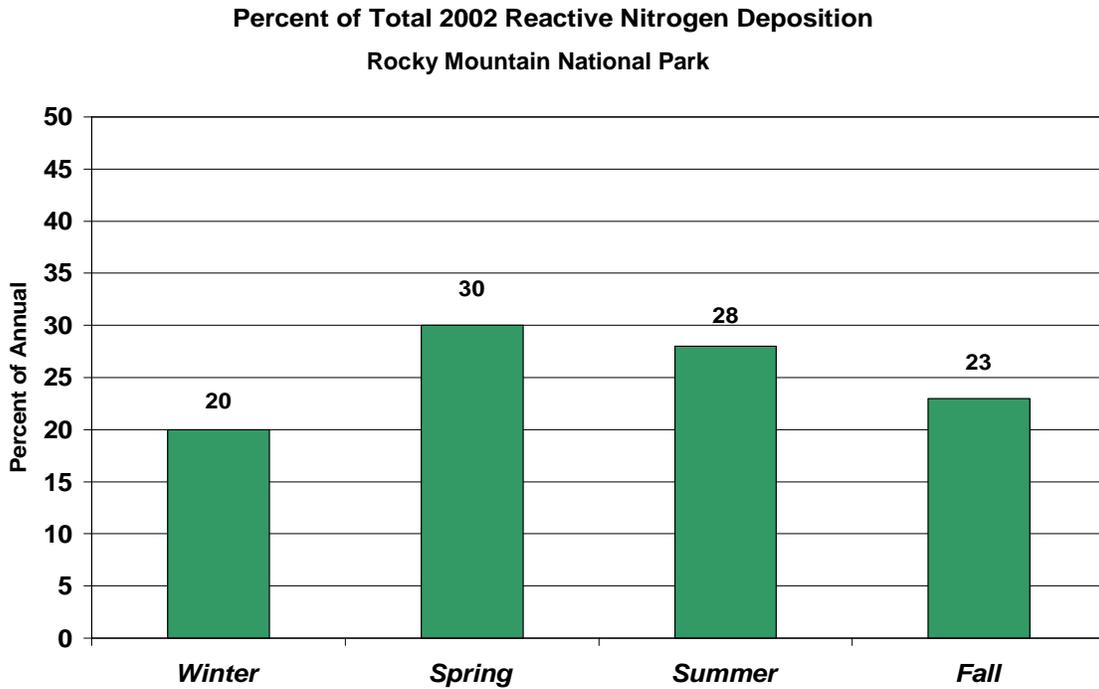
1
2
3

Figure 3.3-6e. Percentage of 2002 total reactive nitrogen deposition in the Shenandoah Case Study Area.

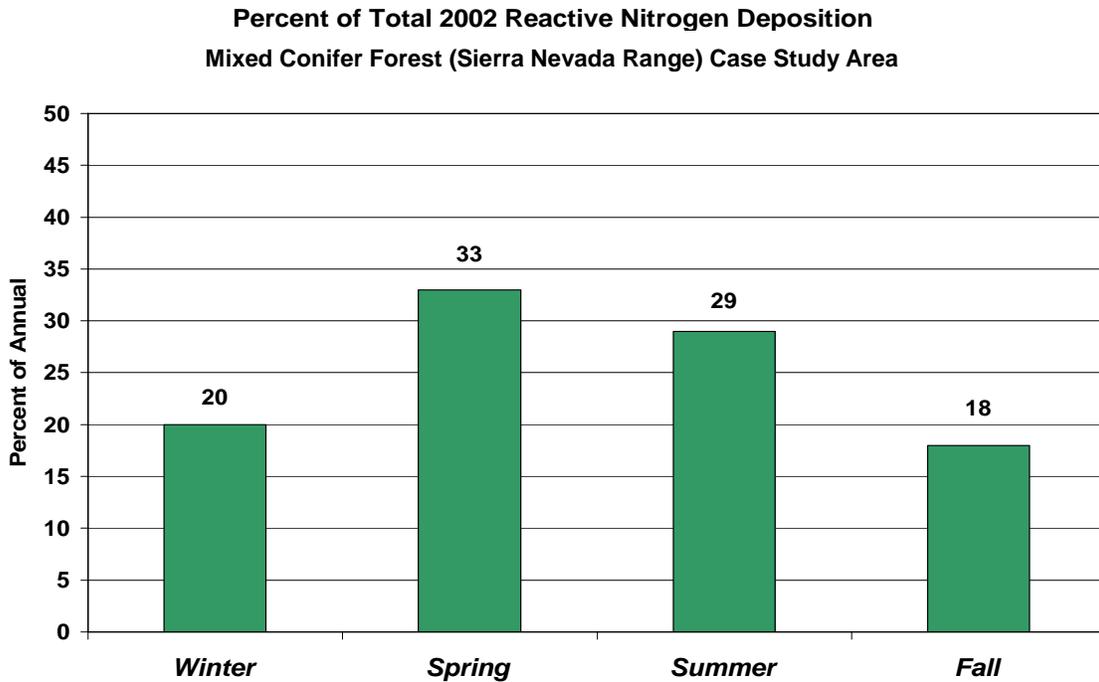


4
5
6

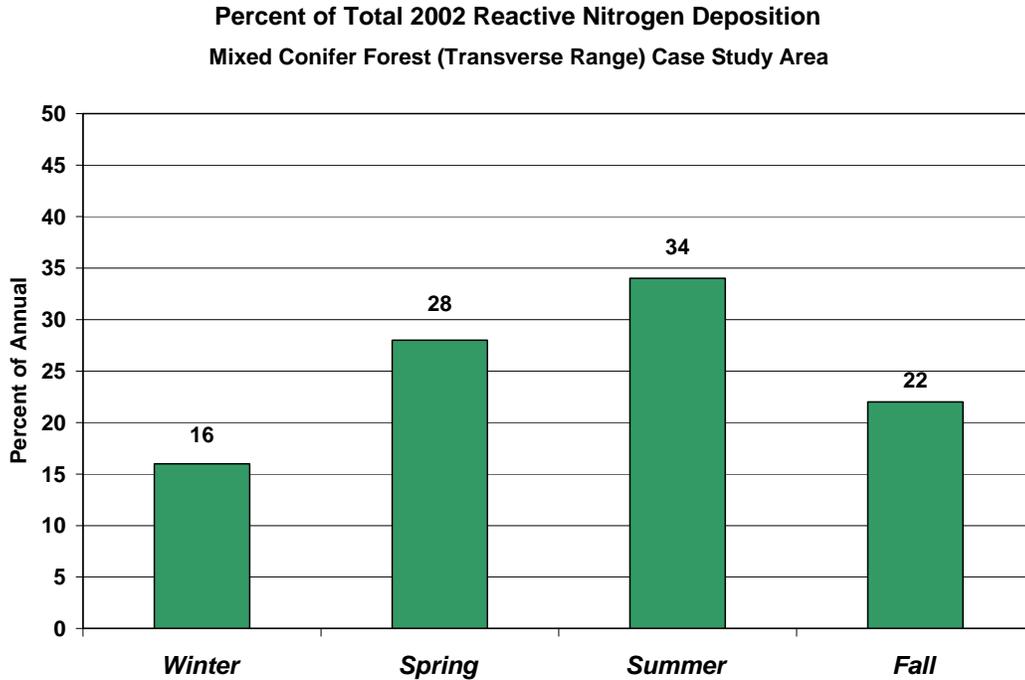
Figure 3.3-6f. Percentage of 2002 total reactive nitrogen deposition in the Neuse River/Neuse River Estuary Case Study Area.



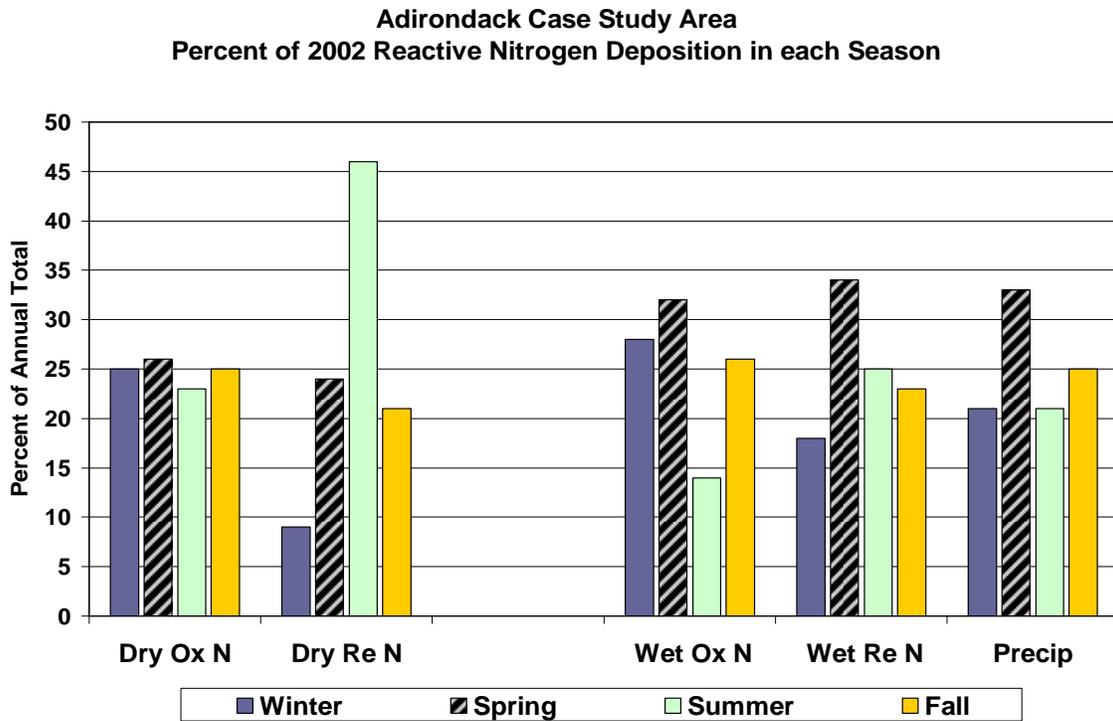
1
2 **Figure 3.3-6g.** Percentage of 2002 total reactive nitrogen deposition in the Rocky
3 Mountain National Park.



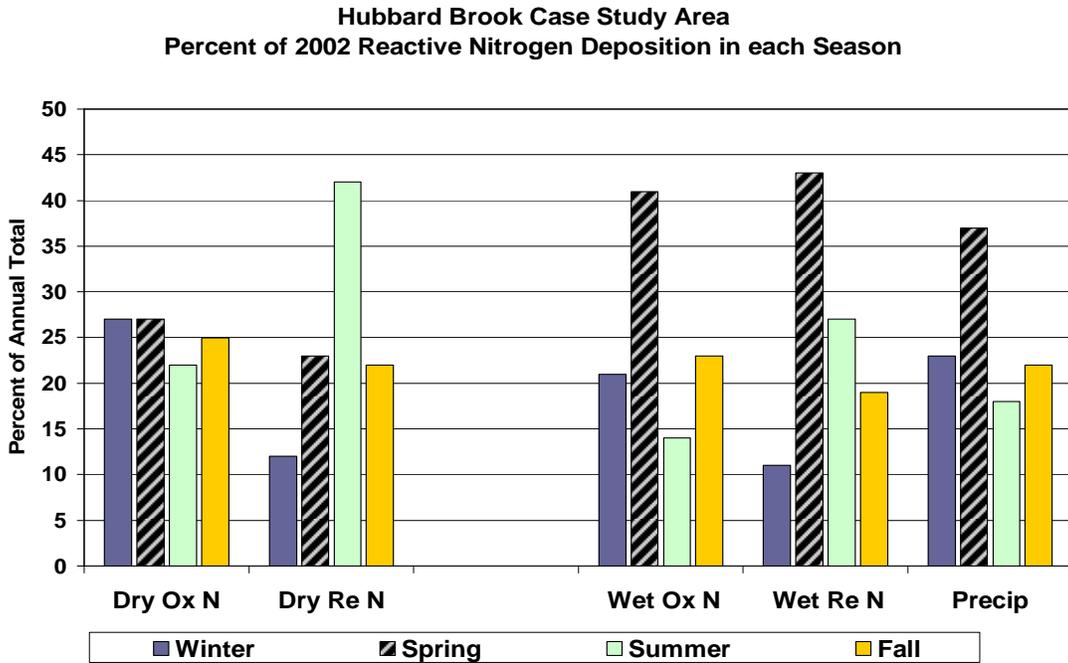
4
5 **Figure 3.3-6h.** Percentage of 2002 total reactive nitrogen deposition in the Sierra
6 Nevada Range portion of the Mixed Conifer Forest Case Study Area.



1
2 **Figure 3.3-6i.** Percentage of 2002 total reactive nitrogen deposition in the
3 Transverse Range portion of the Mixed Conifer Forest Case Study Area.

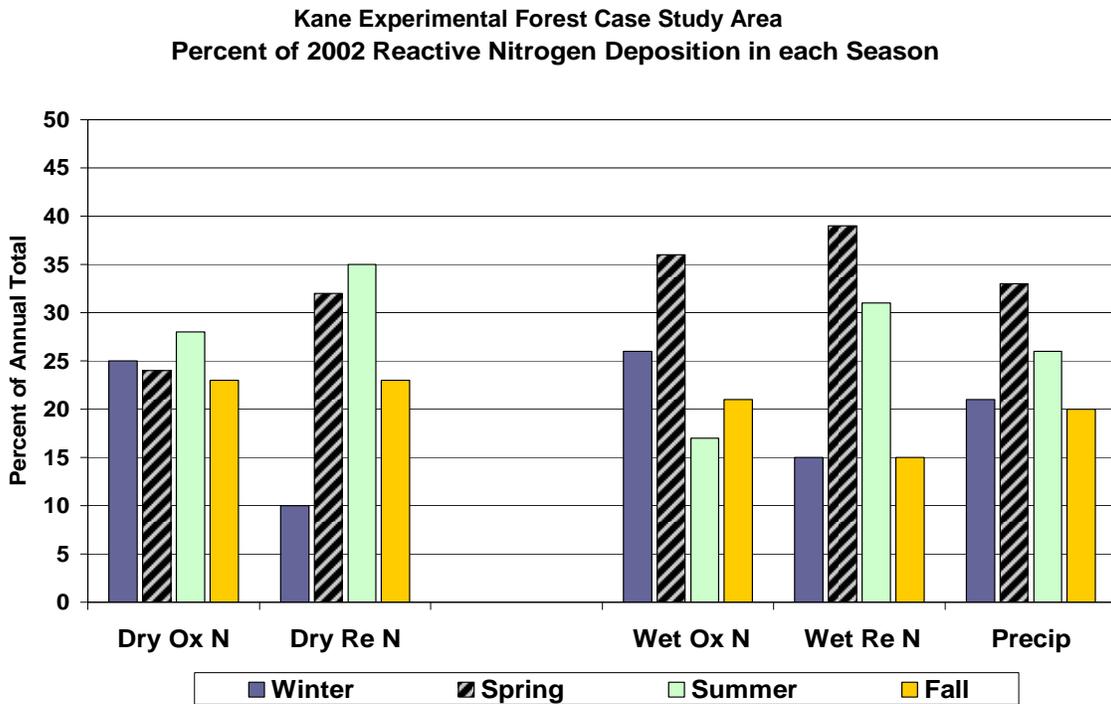


4
5 **Figure 3.3-7a.** Percentage of 2002 reactive nitrogen deposition for each
6 component of nitrogen deposition in the Adirondack Case Study Area.



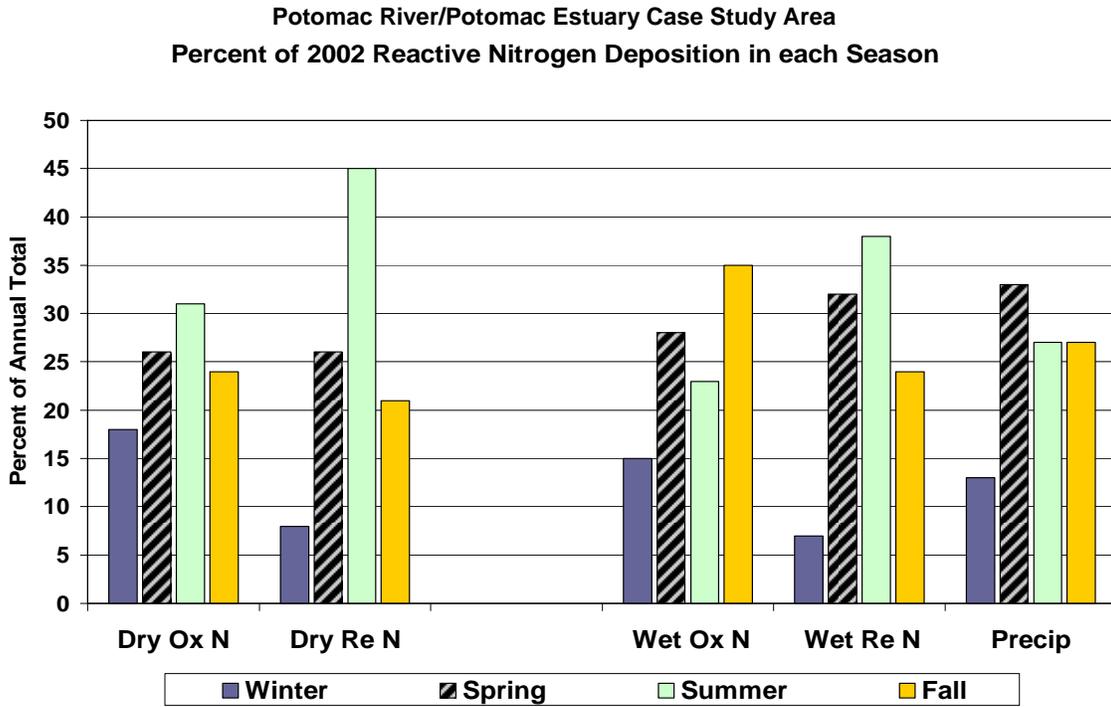
1
2
3
4

Figure 3.3-7b. Percentage of 2002 reactive nitrogen deposition for each component of nitrogen deposition in the Hubbard Brook Experimental Forest Case Study Area.



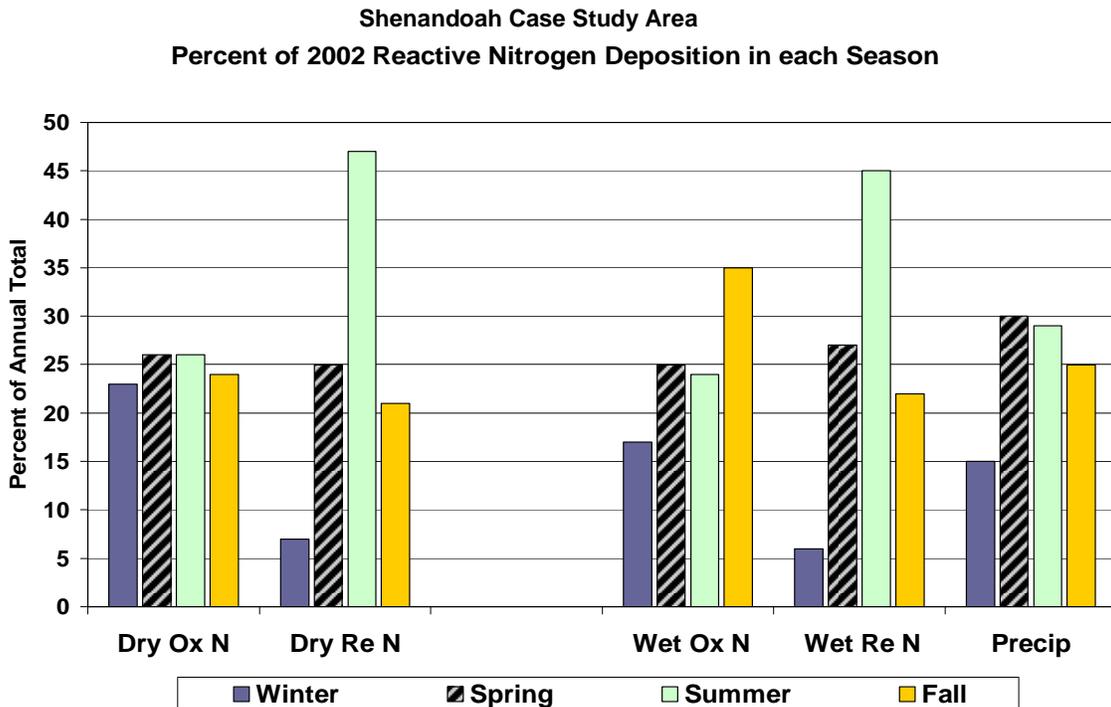
5
6
7
8

Figure 3.3-7c. Percentage of 2002 reactive nitrogen deposition for each component of nitrogen deposition in the Kane Experimental Forest Case Study Area.



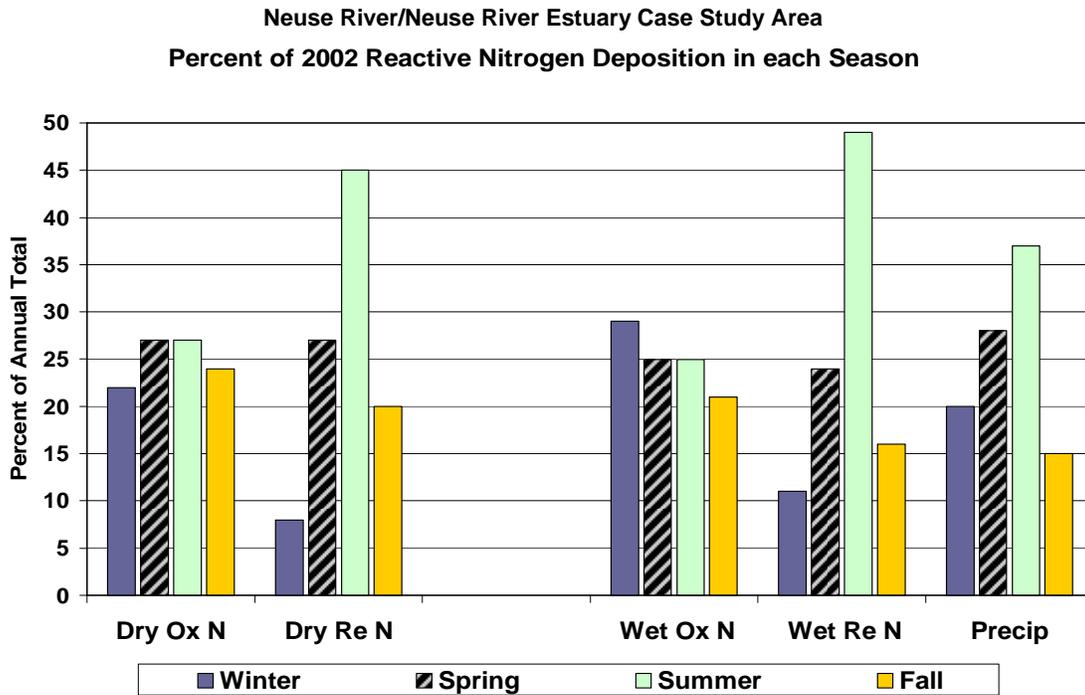
1
2
3
4

Figure 3.3-7d. Percentage of 2002 reactive nitrogen deposition for each component of nitrogen deposition in the Potomac River/Potomac Estuary Case Study Area.



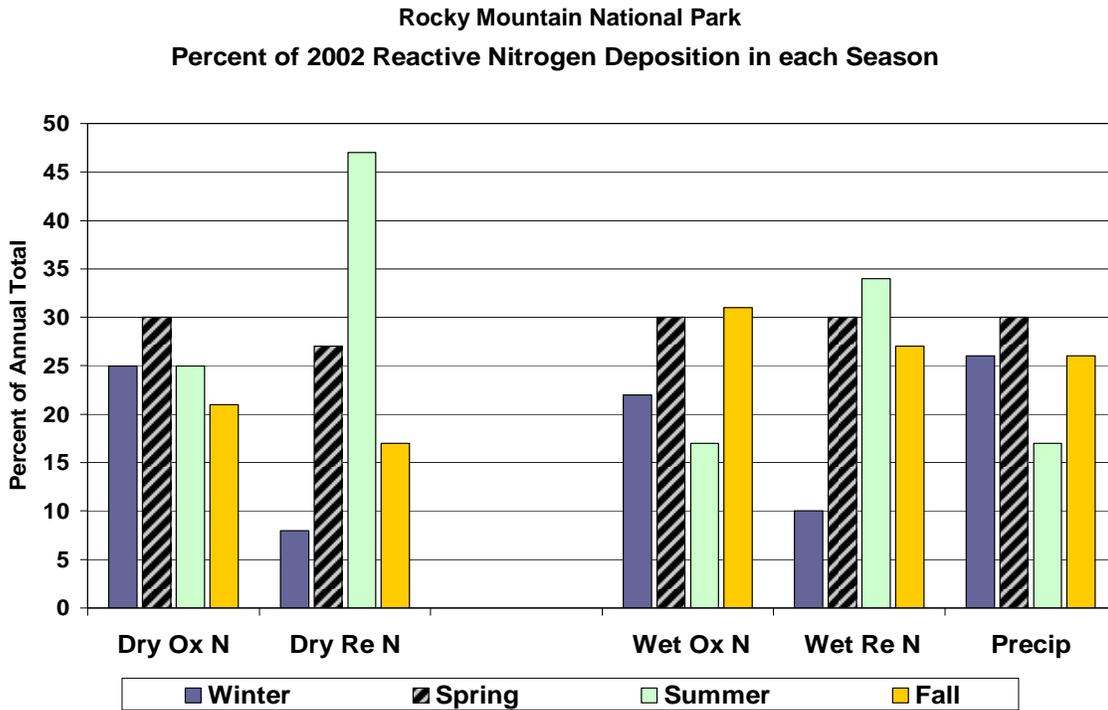
5
6
7

Figure 3.3-7e. Percentage of 2002 reactive nitrogen deposition for each component of nitrogen deposition in the Shenandoah Case Study Area.



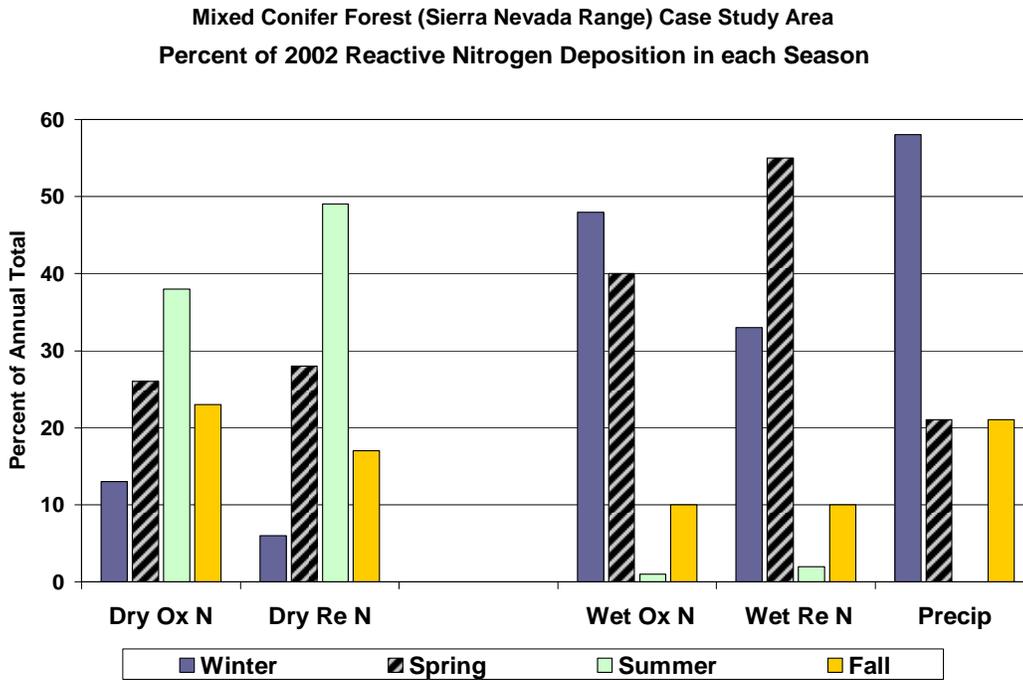
1
2
3
4

Figure 3.3-7f. Percentage of 2002 reactive nitrogen deposition for each component of nitrogen deposition in the Neuse River/Neuse River Estuary Case Study Area.



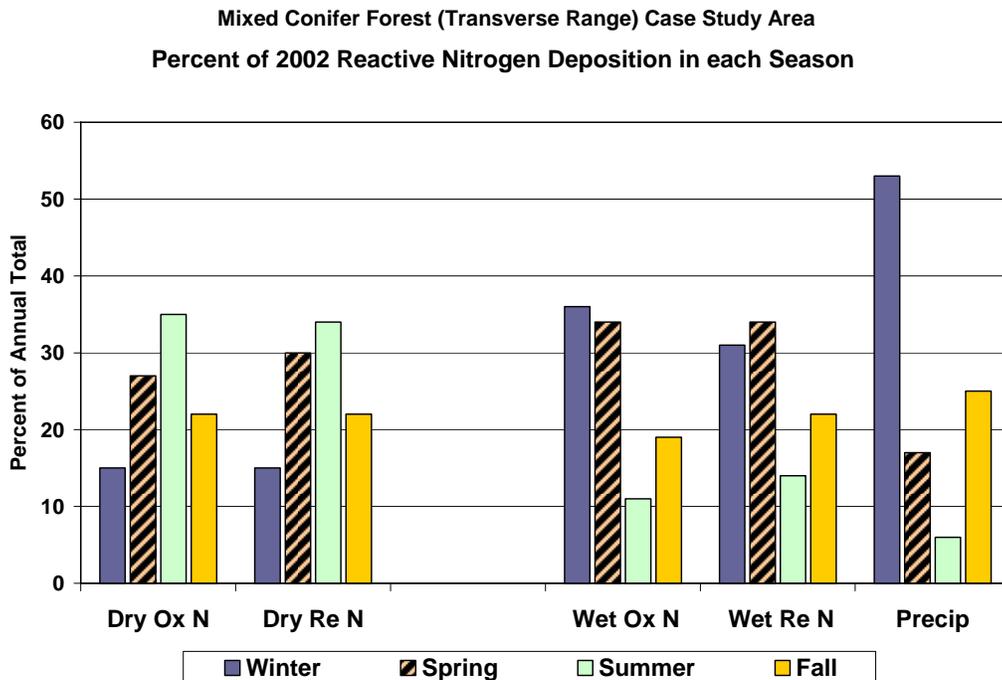
5
6
7

Figure 3.3-7g. Percentage of 2002 reactive nitrogen deposition for each component of nitrogen deposition in the Rocky Mountain National Park.



1
2
3
4

Figure 3.3-7h. Percentage of 2002 reactive nitrogen deposition for each component of nitrogen deposition in the Sierra Nevada Range portion of the Mixed Conifer Forest Case Study Area.



5
6
7
8

Figure 3.3-7i. Percentage of 2002 reactive nitrogen deposition for each component of nitrogen deposition in the Transverse Range portion of the Mixed Conifer Forest Case Study Area.

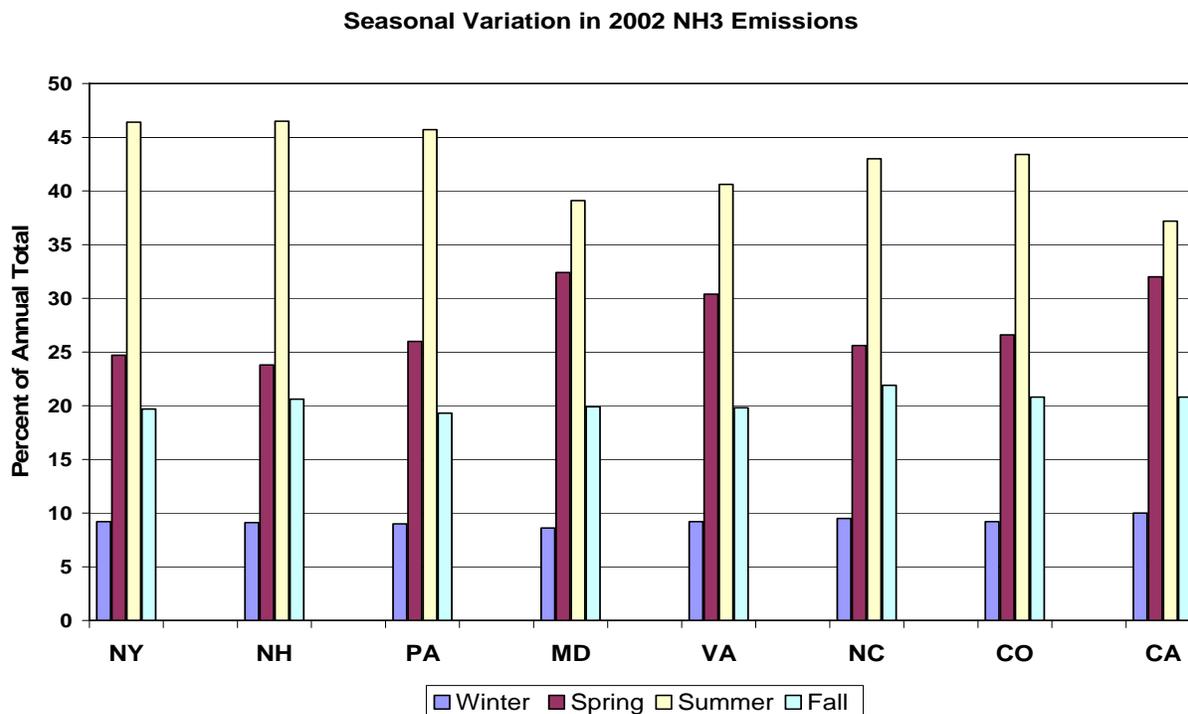


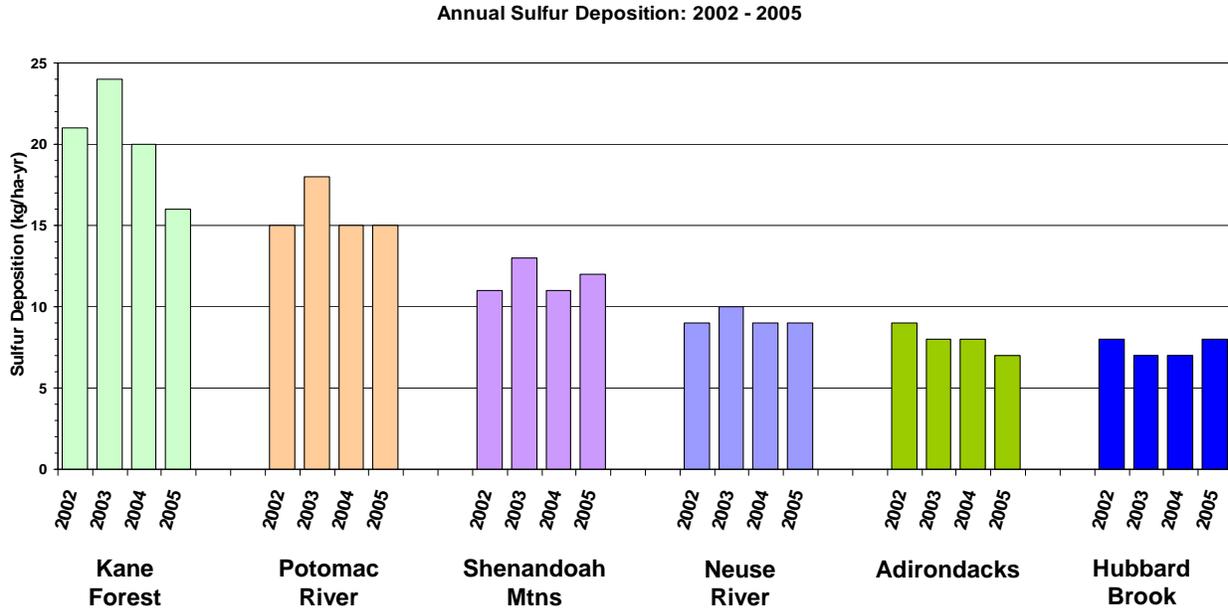
Figure 3.3-8. Percentage of 2002 NH₃ emissions by season for each state containing a case study area.

3.3.3.5 Magnitude of Sulfur Deposition in 2002 and Analysis of Inter-annual Variability

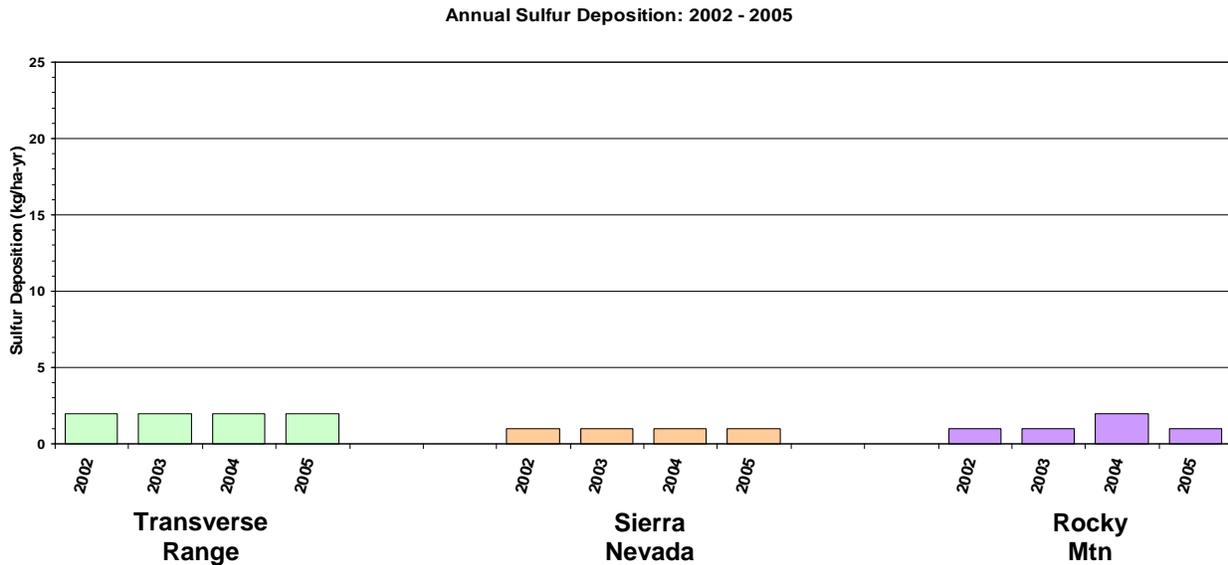
The amount of reactive sulfur deposition in 2002 varies among the case study areas (see **Table 3.3-1**). In the East, sulfur deposition ranges from 7 kg S/ha/yr at the Hubbard Brook Experimental Forest Case Study Area and up to 20 kg S/ha/yr at the Kane Experimental Forest Case Study Area (see **Figure 3.3-9a**). Sulfur deposition in the case study areas in the West is very low and ranged from 1 to 2 kg S/ha/yr (see **Figure 3.3-9b**).

Annual sulfur deposition from 2002 through 2005 varied by 1 to 3 kg S/ha/yr or less at individual case study areas, except for the Kane Experimental Forest Case Study Area, where the range during this period was 8 kg S/ha/yr. There is evidence of a downward trend during this 4-year time period for the Adirondack and Kane Experimental Forest case study areas. No trend is evident during this period for the other case study areas. Trends analyses by CASTNET for an aggregate of 34 sites in the East indicate that dry sulfur deposition levels were fairly steady from 2002 through 2005, followed by a decrease in deposition in 2006 and 2007 (U.S. EPA, 2009). Overall for these 34 sites, sulfur concentrations in wet deposition declined from 2002 to 2004,

1 but then increased from 2005 to 2007 back to the levels monitored in 2002. As in the analysis for
2 nitrogen deposition, trends over the most recent 10-year period were reviewed for wet deposition
3 of sulfur for NADP sites in or near each case study area (see Appendix 2). The site-specific trend
4 information indicates that overall, for each case study area, the amount of sulfur deposition in
5 2002 is generally representative of current conditions. As was found in the analysis of nitrogen
6 deposition, trends in sulfur deposition can vary from site to site, even within a case study area,
7 with the same sites showing high/low amounts of sulfur deposition. In the Adirondack Case
8 Study Area, the data from the Huntington Wildlife Forest site indicate that wet sulfur deposition
9 in 2002 is within the range of values over the most recent 10-year period. However, data from
10 the White Face site show that wet sulfur deposition in 2002 was high compared to that in other
11 years. The data at both sites show a downward trend to 2005, with nitrogen deposition increasing
12 again by 2007. For the Potomac River/Potomac Estuary Case Study Area, the trends in wet
13 sulfur deposition at the Arendtsville, PA, and Parsons, WV, sites indicate that the amount of
14 deposition in 2002 is similar to that from 1998 through 2007. However, the Wye, MD, site on the
15 Eastern Shore of Maryland shows large inter-annual variations compared with the other sites in
16 the Potomac River/Potomac Estuary Case Study Area, and that wet sulfur deposition in 2002 was
17 on the low end of the range over this time period. During the most recent 10-year period, wet
18 sulfur deposition in the two case study areas and Rocky Mountain National Park in the West was
19 low, and generally in the range of 1 to 3 kg S/ha/yr. In 2002, wet sulfur deposition for both the
20 Transverse Range portion of the Mixed Conifer Forest Case Study Area and the Rocky Mountain
21 National Park was at the low end of this range. In the Sierra Nevada Range portion of the Mixed
22 Conifer Forest Case Study Area, wet sulfur deposition in 2002 was within the range of values
23 from 1998 to 2007. Similar to the analysis of nitrogen deposition trends, it was beyond the scope
24 of the current analysis to determine the reasons for the observed trends other than to note that
25 local terrain-induced meteorological conditions and differential source-receptor relationships
26 across a case study area may contribute to the differences noted in deposition trends.



1
2 **Figure 3.3-9a.** Annual sulfur deposition (kg S/ha/yr) from 2002 through 2005 for
3 each case study area in the East.



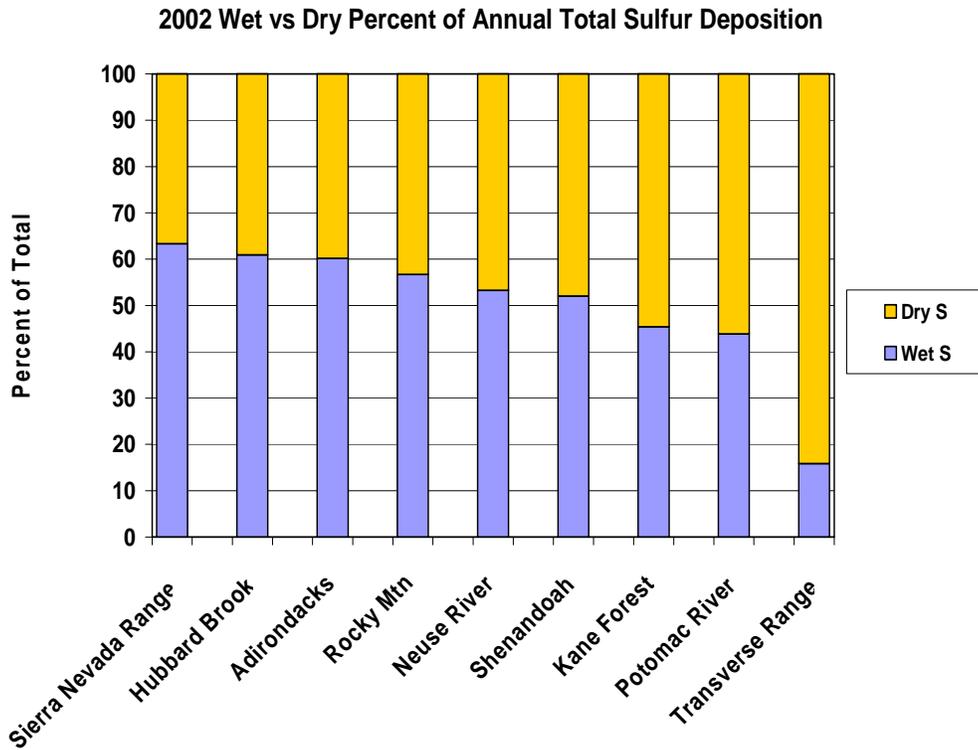
4
5 **Figure 3.3-9b.** Annual sulfur deposition (kg S/ha/yr) from 2002 through 2005 for
6 case study areas in the West, as well as the Rocky Mountain National Park.

7 **3.3.3.6 Relative Amount of Wet and Dry Sulfur Deposition**

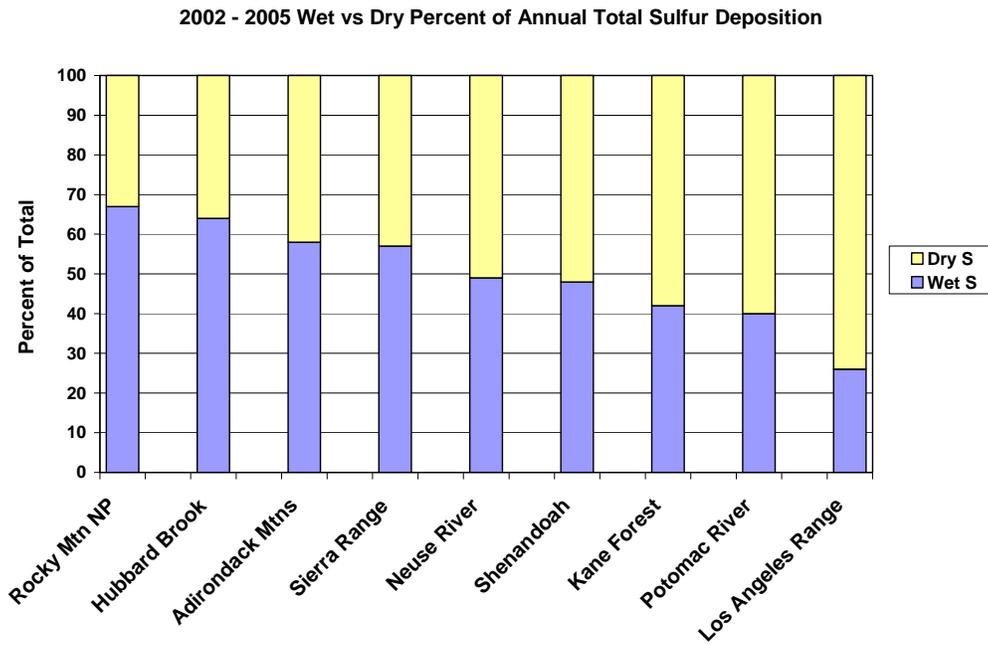
8 The relative amounts of wet and dry sulfur deposition for each case study area are shown
9 in **Figure 3.3-10** for 2002 and in **Figure 3.3-11** for the average of 2002 through 2005. These
10 figures indicate that the relative amounts of wet and dry sulfur deposition in 2002 are consistent

1 with the average over the 4-year time period. The results for the case study areas of the East, as
2 described below, are generally consistent with the findings of Sickles and Shadwick (2007b) on
3 the relative amount of wet and dry sulfur deposition for an aggregation of 34 eastern CASTNET
4 sites. Factors that can influence the relative amounts of wet and dry sulfur deposition in a given
5 location include geographic variations and climatological conditions, which determine the
6 amount of precipitation and transport patterns and the proximity to local sources of SO₂. In
7 general, for the case study areas, those areas that are farthest from sources of high SO₂ emissions
8 tend to have more sulfur deposition from wet deposition than from dry deposition.

9 Approximately 60% of total sulfur deposition in 2002 occurred through wet deposition in the
10 Hubbard Brook Experimental Forest, Adirondack and Mixed Conifer Forest (Sierra Nevada
11 Range portion) case study areas, as well as the Rocky Mountain National Park. Each of these
12 areas is fairly distant from sources of high SO₂ emissions. The relative amounts of wet and dry
13 deposition are about the same in the Shenandoah and Neuse River/Neuse River Estuary case
14 study areas. In the Kane Experimental Forest and Potomac River/Potomac Estuary case study
15 areas, which contain or are close to sources of relatively high SO₂ emissions, dry deposition
16 contributes nearly 60% of the total sulfur deposition. In the Transverse Range portion of the
17 Mixed Conifer Forest Case Study Area, which has a more arid climatology compared with the
18 other areas, >70% of the total sulfur deposition is dry deposited.



1
2 **Figure 3.3-10.** Relative amount of wet and dry annual sulfur deposition in 2002
3 for case study areas.



4
5 **Figure 3.3-11.** Relative amount of wet and dry annual sulfur deposition based on
6 deposition for the period 2002 through 2005 for each case study area and the
7 Rocky Mountain National Park.

1 **3.3.3.7 Geographic Variations in Annual Sulfur Deposition for 2002**²⁵

2 The spatial patterns in total sulfur deposition and wet and dry sulfur deposition in the
3 East are shown in **Figures 3.3-12a-c**. Spatial patterns in total sulfur deposition in the West are
4 shown in **Figure 3.3-13**²⁶.

5 **Adirondack Case Study Area**

6 The highest amounts of sulfur deposition in the Adirondack Case Study Area are found in
7 the southwestern portion of this area, where sulfur deposition is >10 kg S/ha/yr. In the central
8 and eastern sections of this area, sulfur deposition is <8 kg S/ha/yr. Wet deposition of sulfur is
9 greater than dry deposition across all of this area. The spatial gradients in wet sulfur deposition
10 appear to be much stronger than the gradients in dry sulfur deposition. Like nitrogen deposition,
11 the relatively high total sulfur deposition in the southwestern portion of the Adirondack Case
12 Study Area is part of a broad area of high sulfur deposition that stretches along the southern
13 shore of Lake Ontario into western Pennsylvania and beyond.

14 **Shenandoah Case Study Area**

15 The Shenandoah Case Study Area is on the eastern side of the region of high sulfur
16 deposition that covers portions of the Ohio River Valley and West Virginia. Within the
17 Shenandoah Case Study Area, there are several relatively isolated locations with sulfur
18 deposition of >14 kg S/ha/yr. These locations appear to correspond to the location of local
19 sources of high SO₂ emissions, as shown in **Figure 3.2-5**. There is a large range in dry sulfur
20 deposition within the Shenandoah Case Study Area, with amounts ranging from 3 to 4 kg S/ha/yr
21 up to 14 kg S/ha/yr. Wet sulfur deposition appears to be spatially more homogeneous than dry
22 sulfur deposition. Amounts of wet sulfur deposition range from 5 to 6 kg S/ha/yr across most of
23 the area, with higher amounts, up to the range of 6 to 7 kg S/ha/yr, found in the northwestern part
24 of the area.

25 **Potomac River/Potomac Estuary Case Study Area**

26 There was considerable variation in sulfur deposition across the Potomac River/Potomac
27 Estuary Case Study Area in 2002. The highest amounts of sulfur deposition in this area, of 24 kg

²⁵ Note that an analysis of the spatial gradients in reactive nitrogen and sulfur deposition for the Kane Experimental Forest and Hubbard Brook Experimental Forest case study areas, as well as the Rocky Mountain National Park, is not included because the size of each of these areas is small relative to the 12 × 12-km resolution-measured data and model predictions used in this analysis.

²⁶ See footnote 19 for caveats concerning the analysis of geographic variations in deposition for the case study areas in the West.

1 S/ha/yr or more, are found in the far northwestern portion of this area, which is near sources of
2 high SO₂ emissions in western Pennsylvania. Lower amounts of sulfur deposition of 14 kg
3 S/ha/yr or more is found over the eastern half of the Potomac River/Potomac Estuary Case Study
4 Area. The lowest amount of sulfur deposition, in the range of 8 to 10 kg S/ha/yr, is seen in the far
5 southwest portion of this area. Wet and dry sulfur depositions are both relatively high in the
6 northwestern portion of this area. In the eastern portion of this area, near the sources of SO₂
7 emissions in the vicinity of Washington, DC, dry sulfur deposition is greater than wet.

8 **Neuse River/Neuse River Estuary Case Study Area**

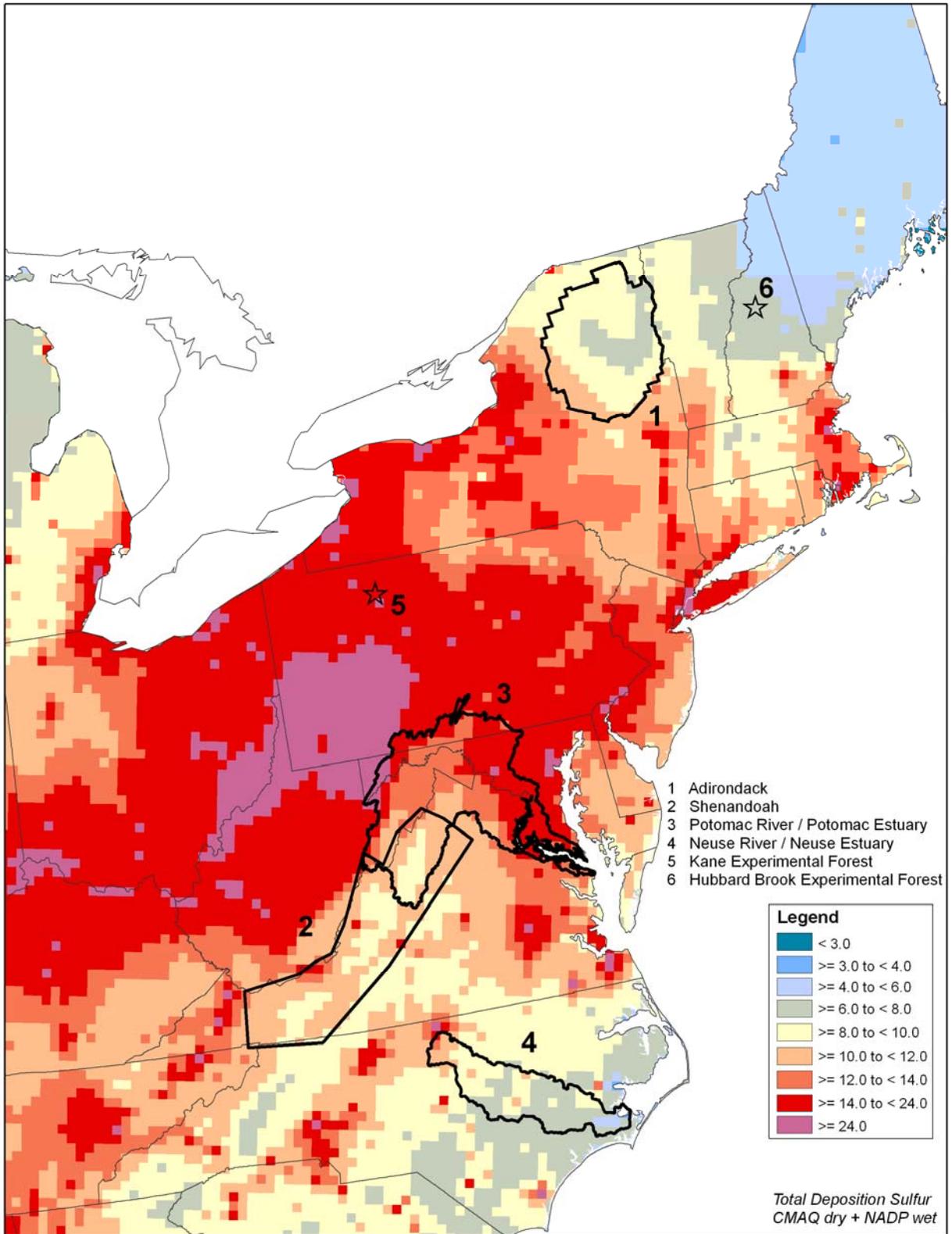
9 In the Neuse River/Neuse River Estuary Case Study Area, sulfur deposition is highest
10 near the Raleigh-Durham urban area (14 kg S/ha/yr or more), and in particular, near a source of
11 high SO₂ emissions located near the North Carolina/Virginia border. Sulfur deposition generally
12 decreases from northwest to southeast down to 6 to 8 kg S/ha/yr in the eastern portion of this
13 area. Most of the spatial variation in sulfur deposition appears to be associated with dry
14 deposition. Dry sulfur deposition increases from 2 to 3 kg S/ha/yr near the mouth of the Neuse
15 River up to 9 to 14 kg S/ha/yr in the northwest corner of this case study area. In contrast, the
16 amount of wet sulfur deposition appears to be fairly homogeneous across most of the case study
17 area, with amounts in the range of 4 to 5 kg S/ha/yr.

18 **Sierra Nevada Range (a Portion of the Mixed Conifer Forest Case Study Area)**

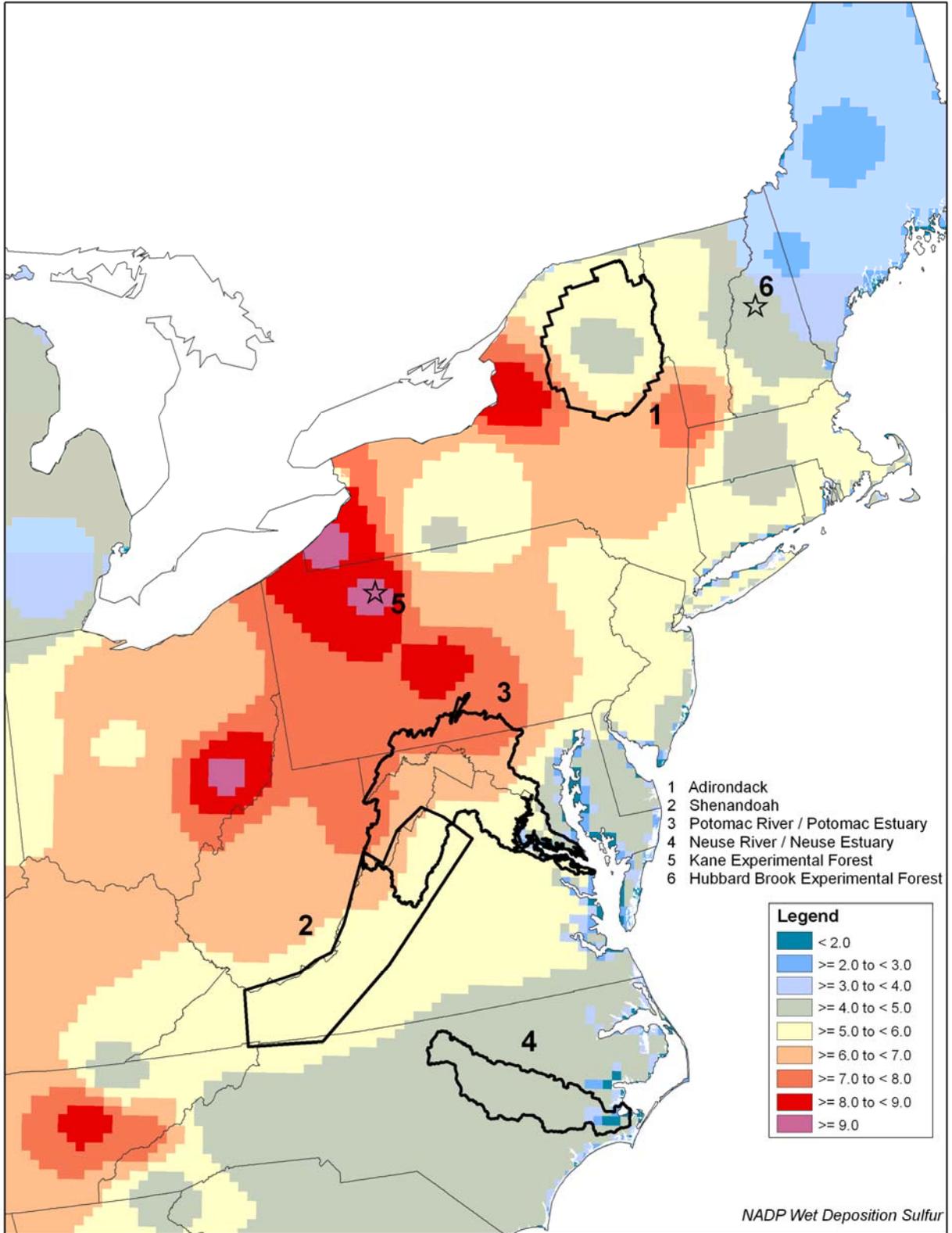
19 There appears to be very little spatial variation in sulfur deposition in the Sierra Nevada
20 Range portion of the Mixed Conifer Forest Case Study Area. The amount of sulfur deposition is
21 <1 kg S/ha/yr across most of this area. The highest amounts (1 to 2 kg S/ha/yr) are found in the
22 extreme western portion of this area.

23 **Transverse Range (a Portion of the Mixed Conifer Forest Case Study Area)**

24 In the Transverse Range portion of the Mixed Conifer Forest Case Study Area, sulfur
25 deposition decreases with distance from the Los Angeles urban area. Sulfur deposition in the San
26 Bernardino Mountains north of Los Angeles is in the range of 0.5 to 2 kg S/ha/yr.

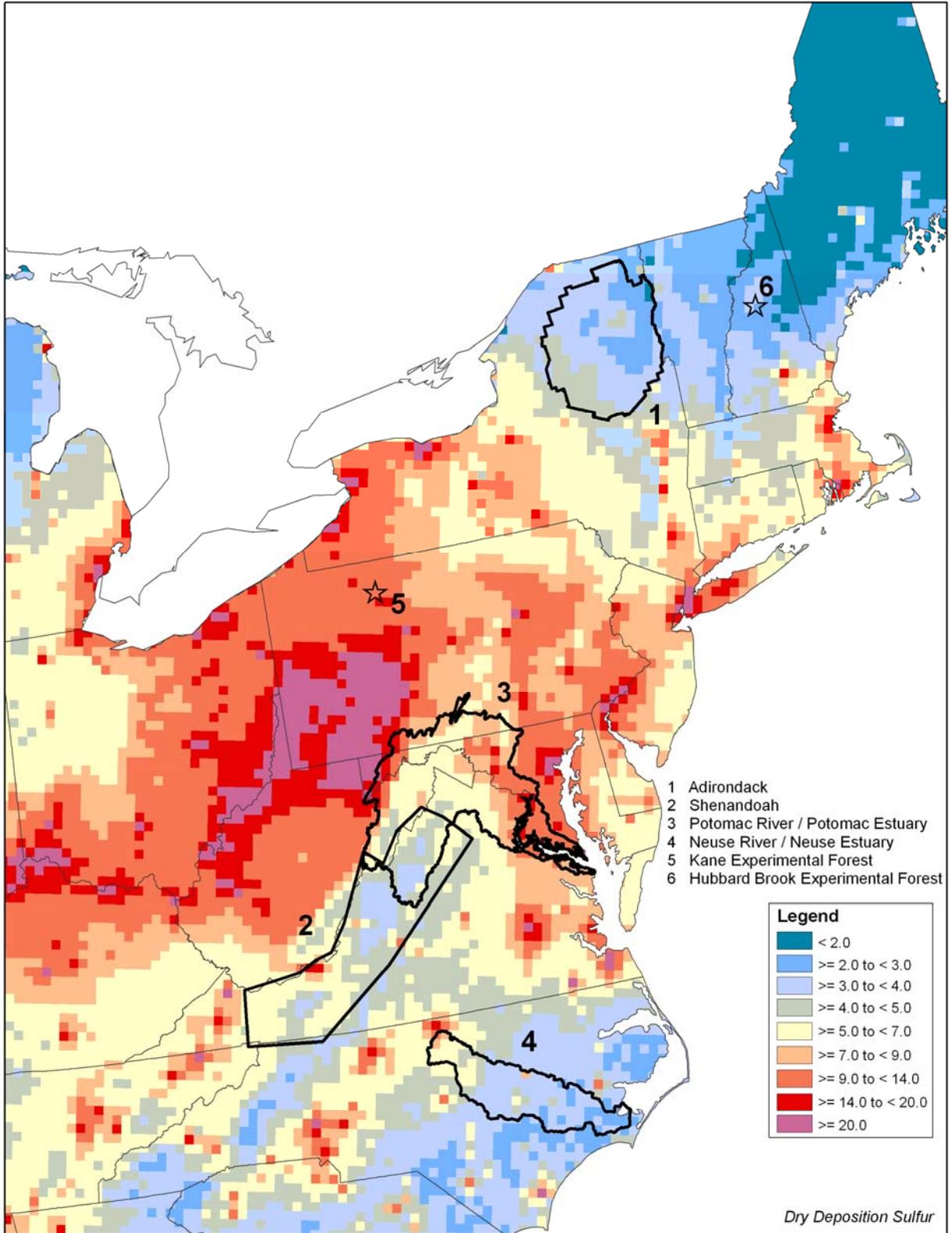


1
 2 **Figure 3.3-12a.** Annual total dry plus wet sulfur deposition (kg S/ha/yr) in 2002
 3 for the case study areas in the East.



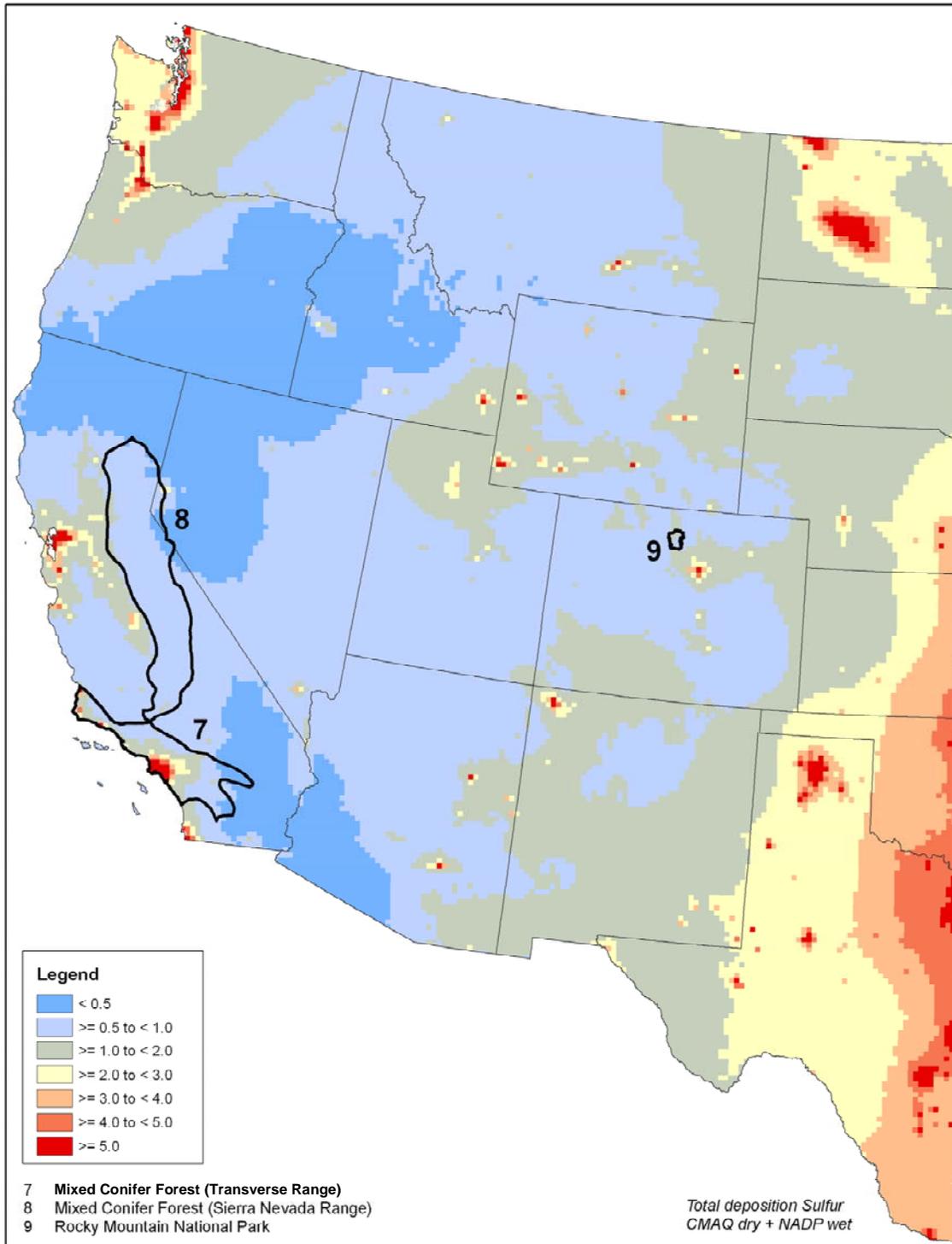
1
 2
 3

Figure 3.3-12b. Annual wet sulfur deposition (kg S/ha/yr) in 2002 for the case study areas in the East.



1
 2
 3

Figure 3.3-12c. Annual dry sulfur deposition (kg S/ha/yr) in 2002 for the case study areas in the East.



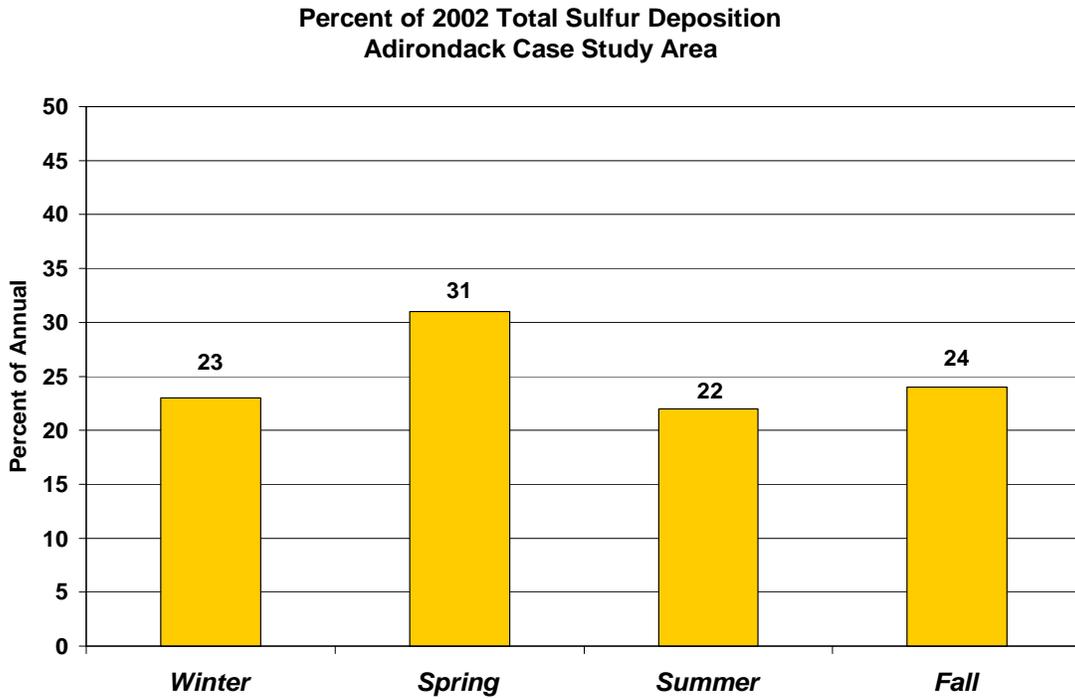
1
2
3

Figure 3.3-13. Annual total dry plus wet sulfur deposition (kg S/ha/yr) in 2002 for case study areas and Rocky Mountain National Park in the West.

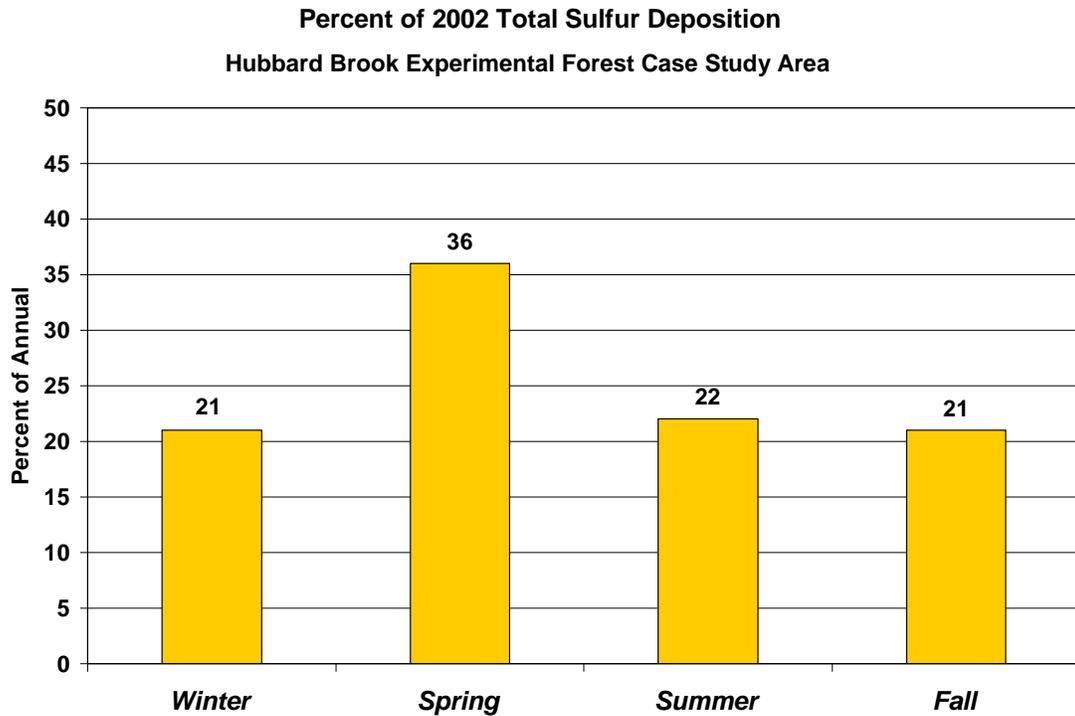
1 **3.3.3.8 Seasonal Variations in Sulfur Deposition for 2002**

2 The seasonal patterns in total sulfur deposition for each case study area are shown in
3 **Figures 3.3-14(a-i)**, and the seasonal patterns for wet and dry sulfur deposition and precipitation
4 are shown in **Figures 3.3-15(a-i)**. Sulfur deposition is greatest in spring or summer, except in the
5 Sierra Nevada Range portion of the Mixed Conifer Forest Case Study Area, as described below.
6 For the case study areas of the East, the seasonal patterns in sulfur deposition are generally
7 similar to those of total reactive nitrogen deposition. Thus, these areas are affected by the highest
8 amount of sulfur deposition and total reactive nitrogen deposition during the same season.
9 Examination of the seasonal variations in wet and dry sulfur deposition in the case study areas in
10 the East indicates that dry sulfur deposition is highest in winter and lowest in summer, whereas
11 wet sulfur deposition peaks in spring or summer and generally tracks the seasonal patterns in
12 precipitation.

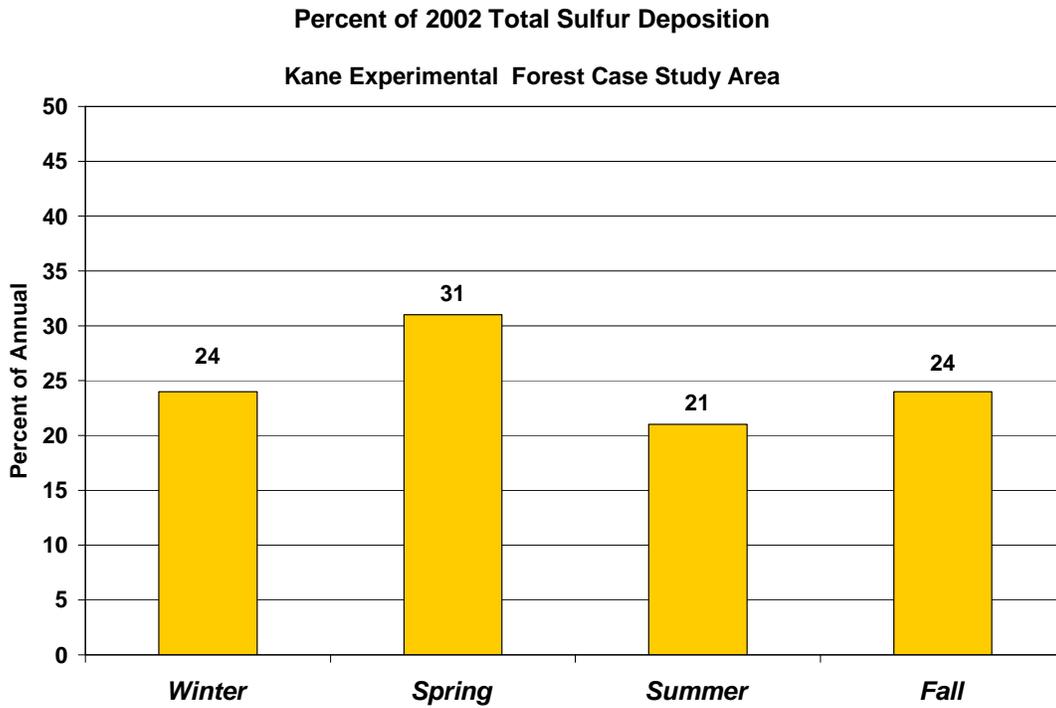
13 In the case study areas in the West, the seasonal patterns in wet sulfur deposition are very
14 similar to the precipitation patterns that were found for the case study areas in the East. In the
15 Sierra Nevada Range and Transverse Range (Mixed Conifer Forest Case Study Area), there are
16 large seasonal variations in precipitation, which affect the seasonal variations in wet sulfur
17 deposition. In these two areas, nearly all of the wet sulfur deposition occurs during winter and
18 spring, which are the seasons with the most of the precipitation. The seasonal patterns in total
19 sulfur deposition reflect the net effect of the seasonal variations in wet and dry sulfur deposition.
20 In the Rocky Mountain National Park and the Transverse Range portion of the Mixed Conifer
21 Forest Case Study Area, total sulfur deposition peaks in spring. In contrast, in the Sierra Nevada
22 Range portion of the Mixed Conifer Forest Case Study Area, both winter and spring have much
23 higher sulfur deposition compared with summer and fall.



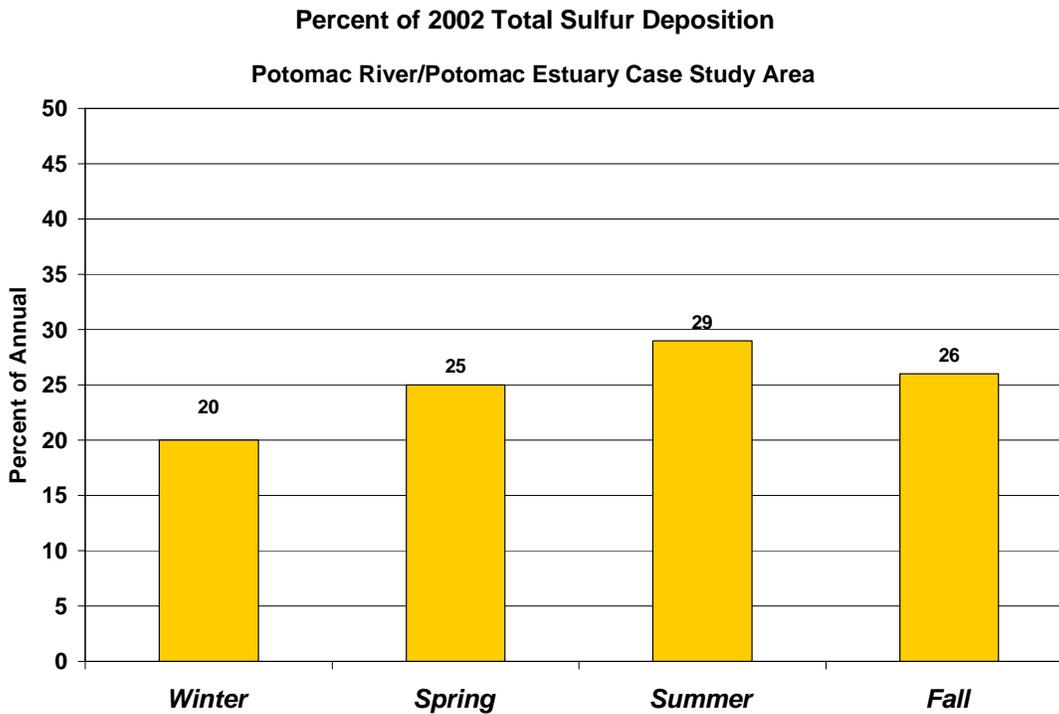
1
2 **Figure 3.3-14a.** Percentage of 2002 total sulfur deposition in the Adirondack Case
3 Study Area.



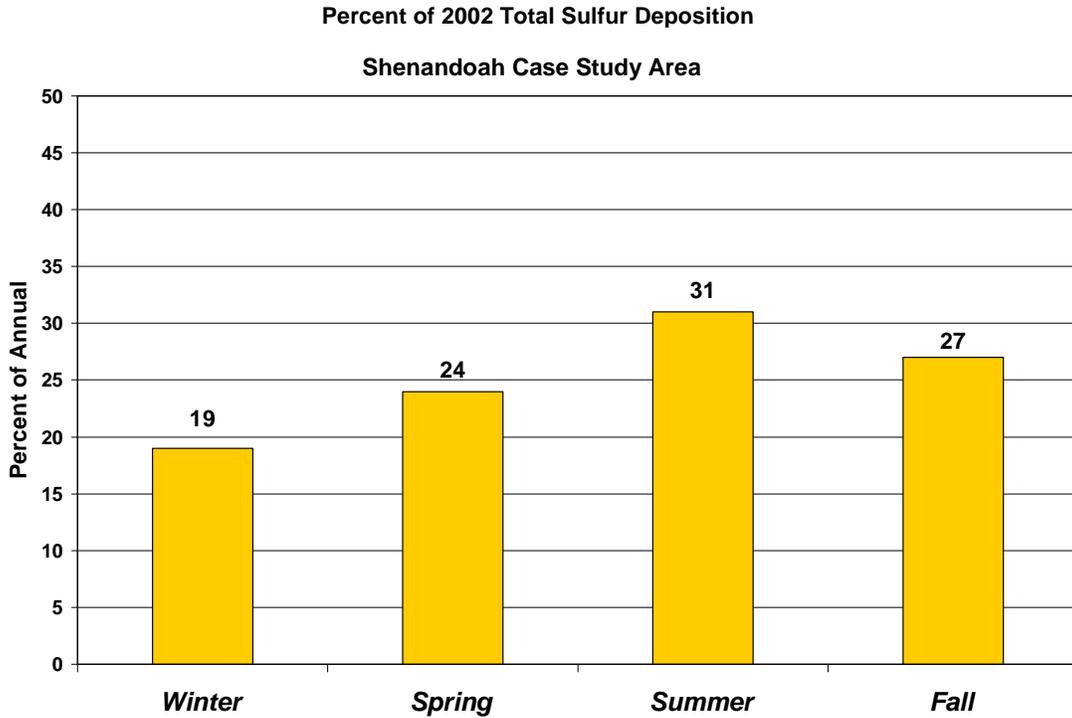
4
5 **Figure 3.3-14b.** Percentage of 2002 total sulfur deposition in the Hubbard Brook
6 Experimental Forest Case Study Area.



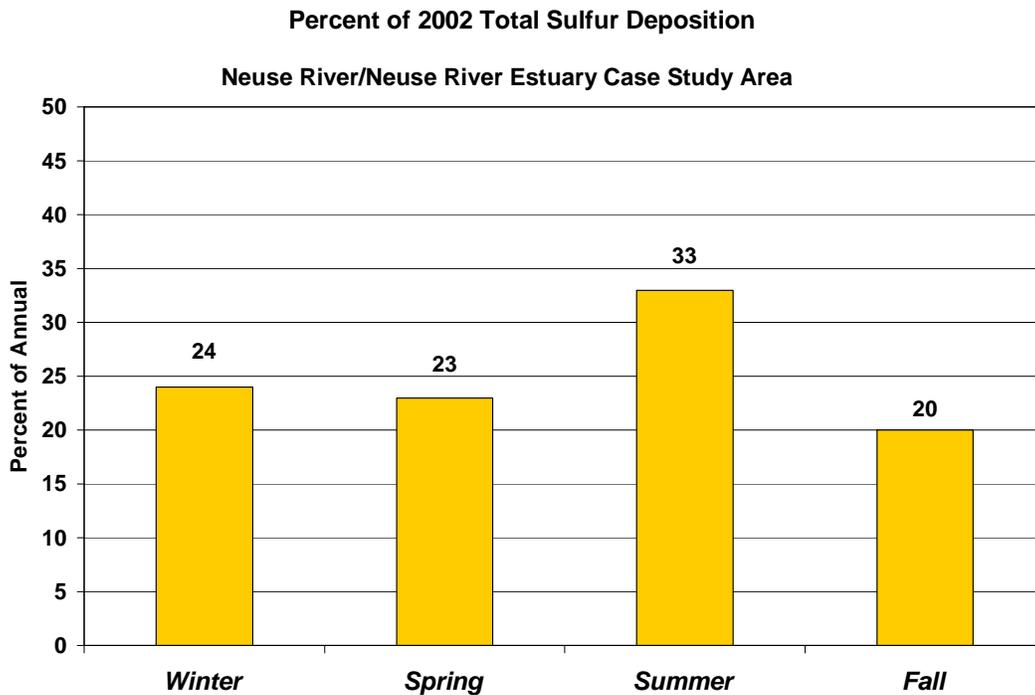
1
2 **Figure 3.3-14c.** Percentage of 2002 total sulfur deposition in the Kane Experimental
3 Forest Case Study Area.



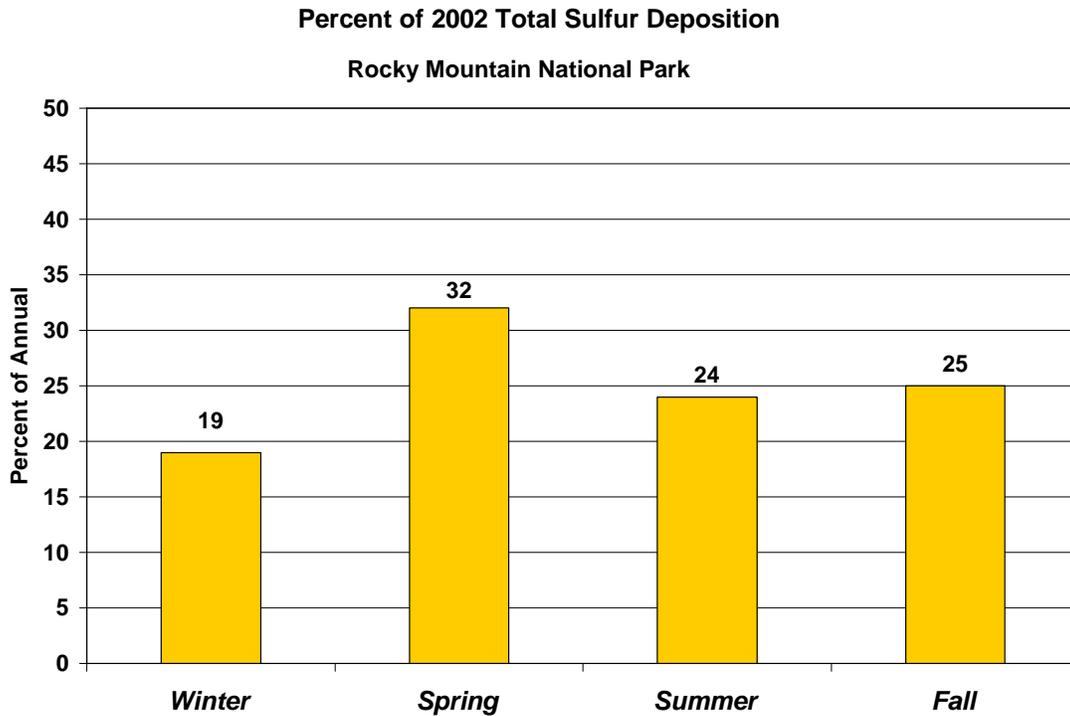
4
5 **Figure 3.3-14d.** Percentage of 2002 total sulfur deposition in the Potomac
6 River/Potomac Estuary Case Study Area.



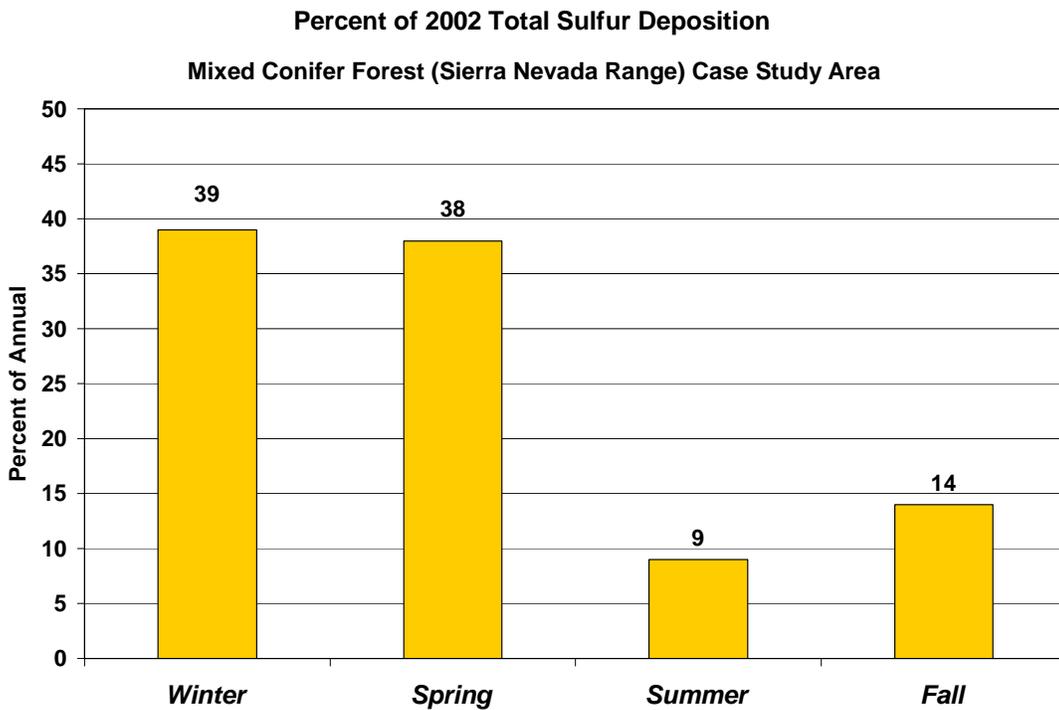
1
2 **Figure 3.3-14e.** Percentage of 2002 total sulfur deposition in the Shenandoah Case
3 Study Area.



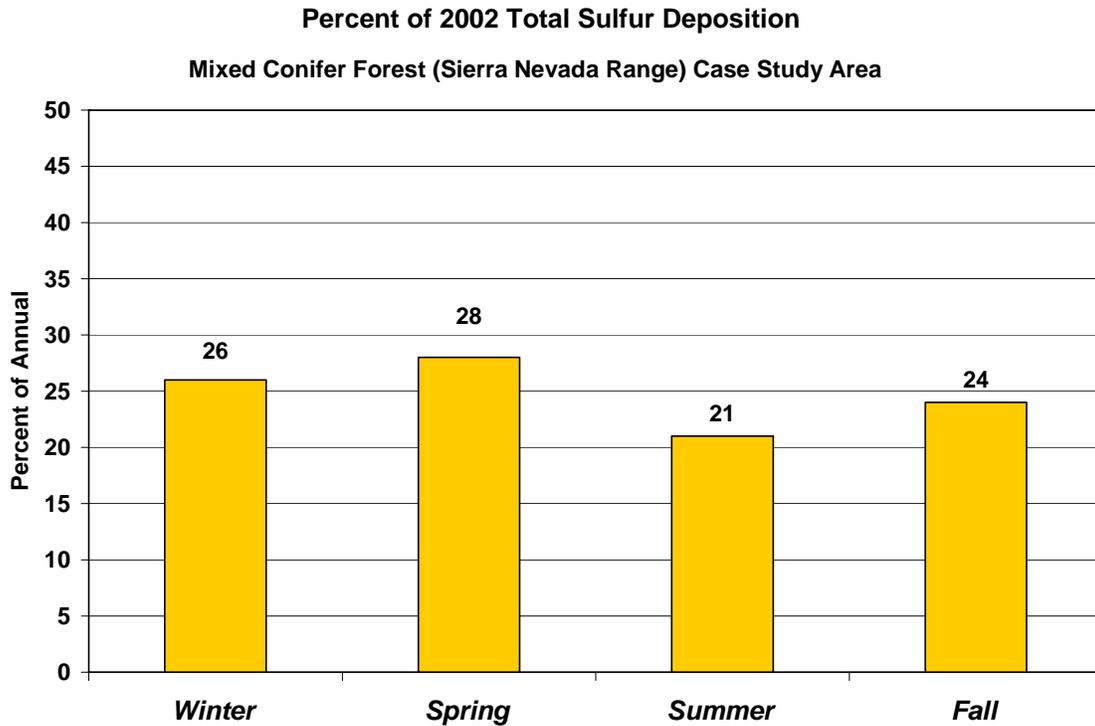
4
5 **Figure 3.3-14f.** Percentage of 2002 total sulfur deposition in the Neuse River/Neuse
6 River Estuary Case Study Area.



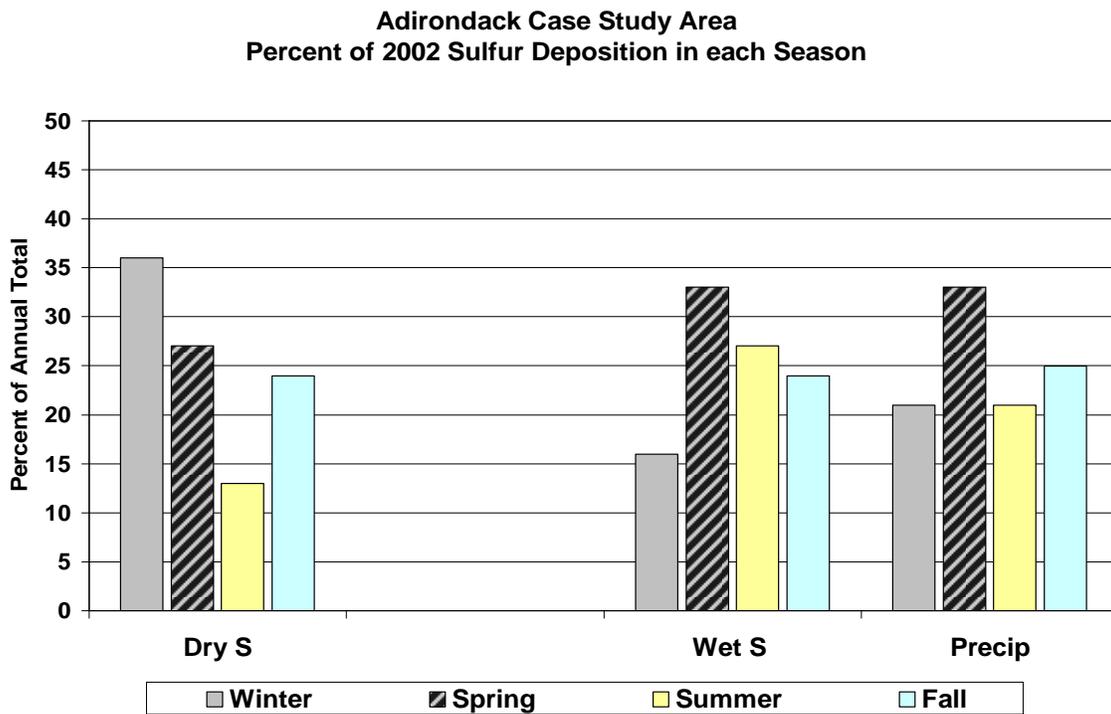
1
2 **Figure 3.3-14g.** Percentage of 2002 total sulfur deposition in the Rocky Mountain
3 National Park.



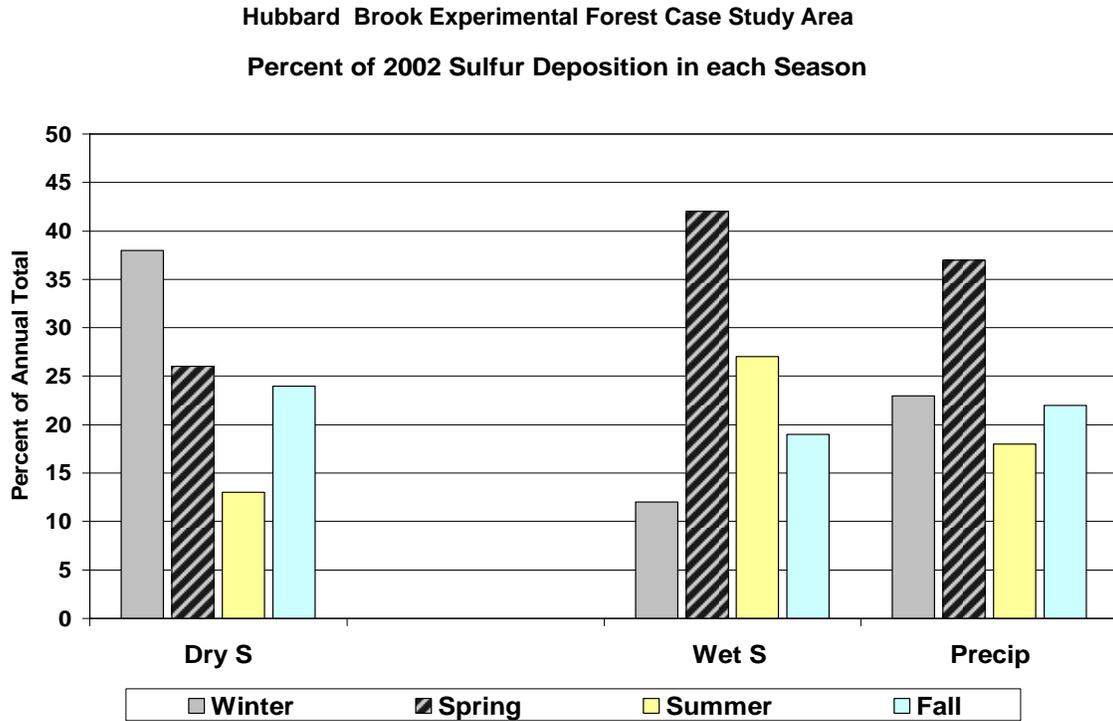
4
5 **Figure 3.3-14h.** Percentage of 2002 total sulfur deposition in the Sierra Nevada
6 Range portion of the Mixed Conifer Forest Case Study Area.



1
 2 **Figure 3.3-14i.** Percentage of 2002 total sulfur deposition in the Transverse Range
 3 portion of the Case Study Area.

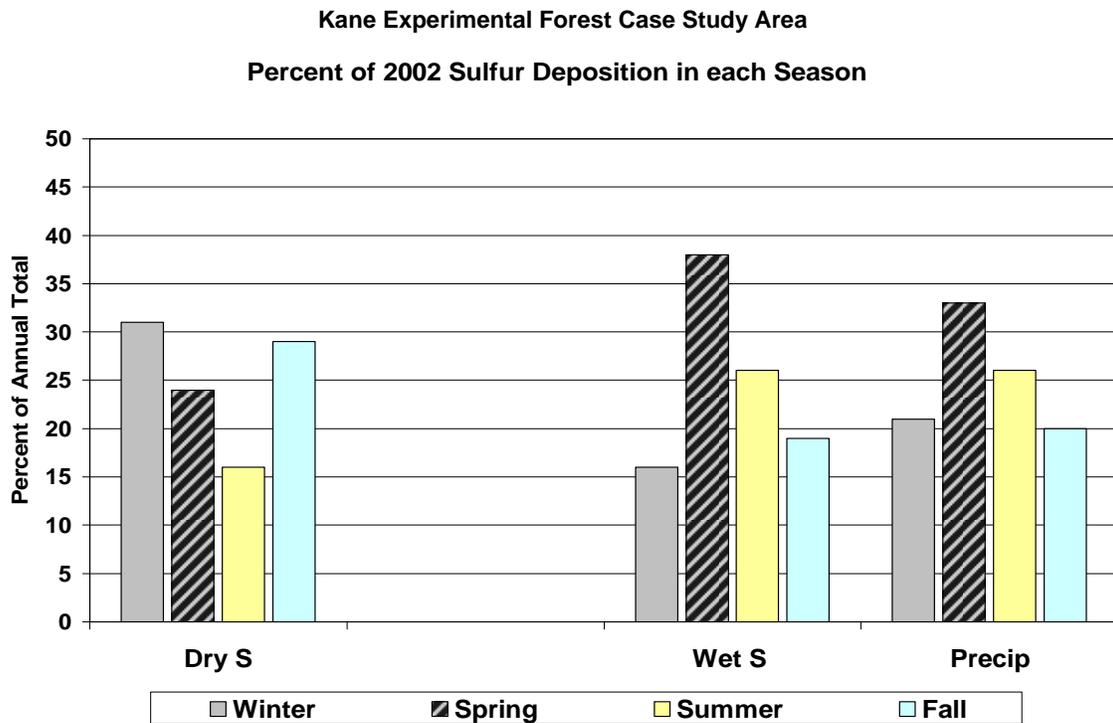


4
 5 **Figure 3.3-15a.** Percentage of 2002 deposition for each component of sulfur
 6 deposition in the Adirondack Case Study Area.



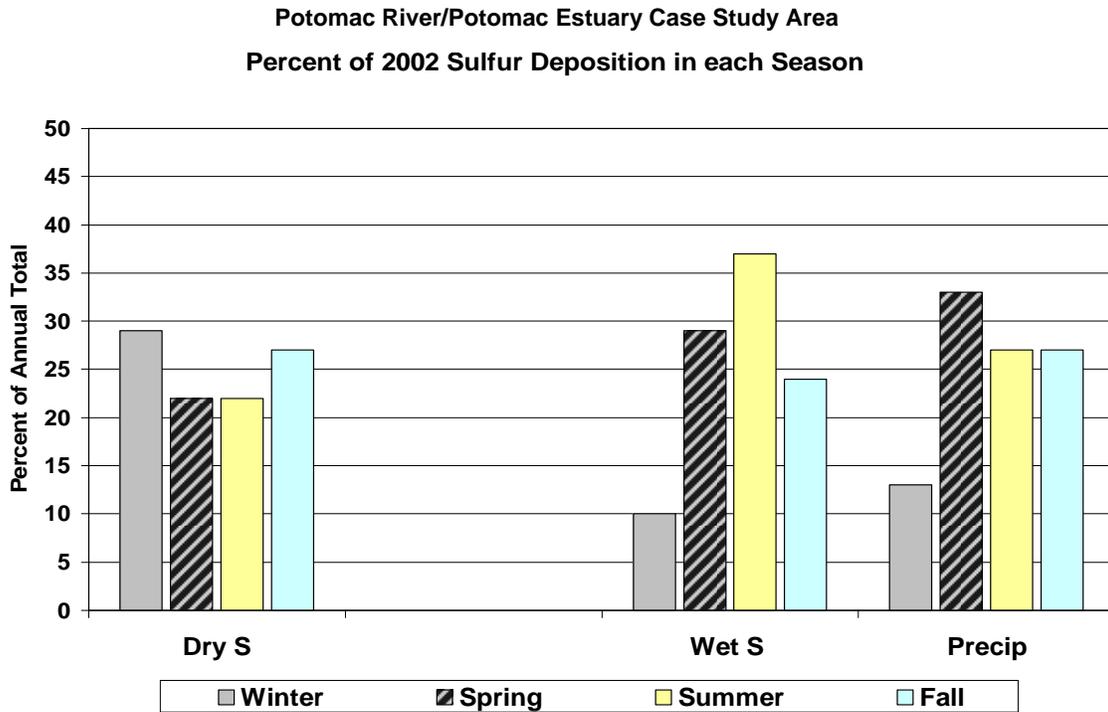
1
2
3

Figure 3.3-15b. Percentage of 2002 deposition for each component of sulfur deposition in the Hubbard Brook Experimental Forest Case Study Area.



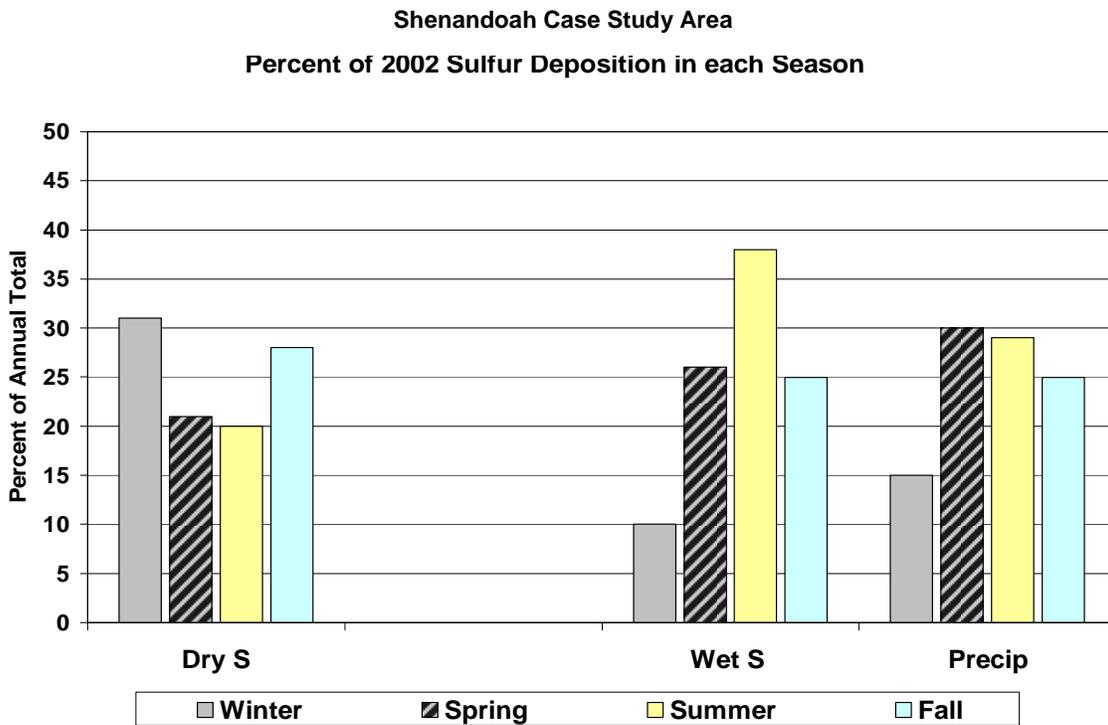
4
5
6

Figure 3.3-15c. Percentage of 2002 deposition for each component of sulfur deposition in the Kane Experimental Forest Case Study Area.



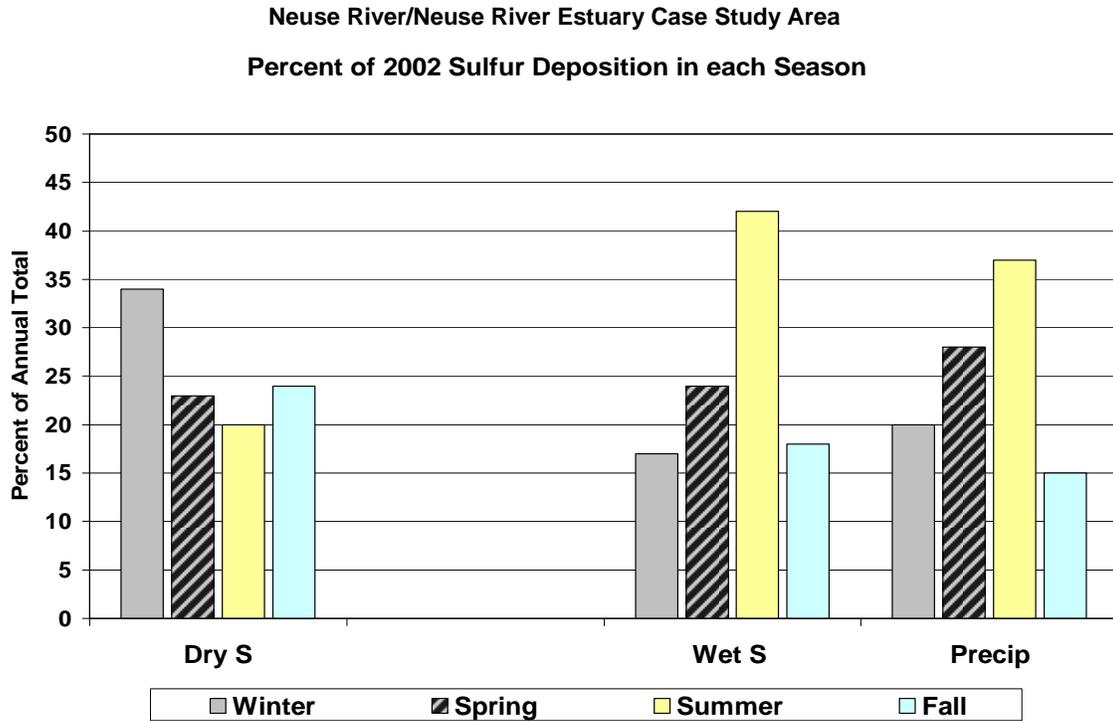
1
2
3

Figure 3.3-15d. Percentage of 2002 deposition for each component of sulfur deposition in the Potomac River/Potomac Estuary Case Study Area.



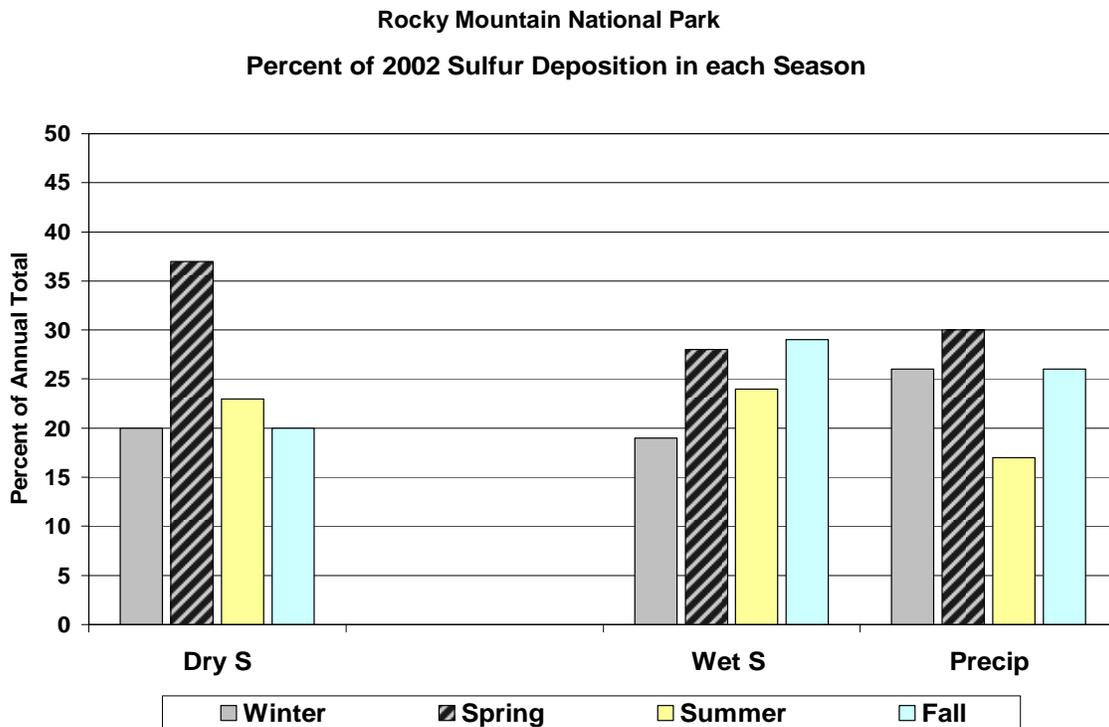
4
5
6

Figure 3.3-15e. Percentage of 2002 deposition for each component of sulfur deposition in the Shenandoah Case Study Area.



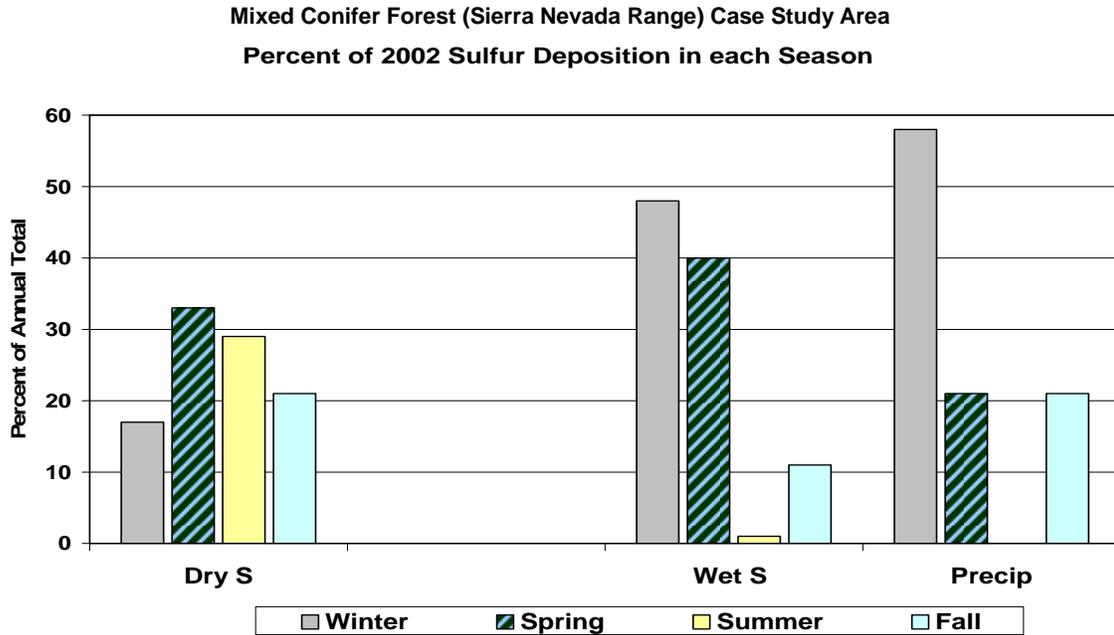
1
2
3

Figure 3.3-15f. Percentage of 2002 deposition for each component of sulfur deposition in the Neuse River/Neuse River Estuary Case Study Area.

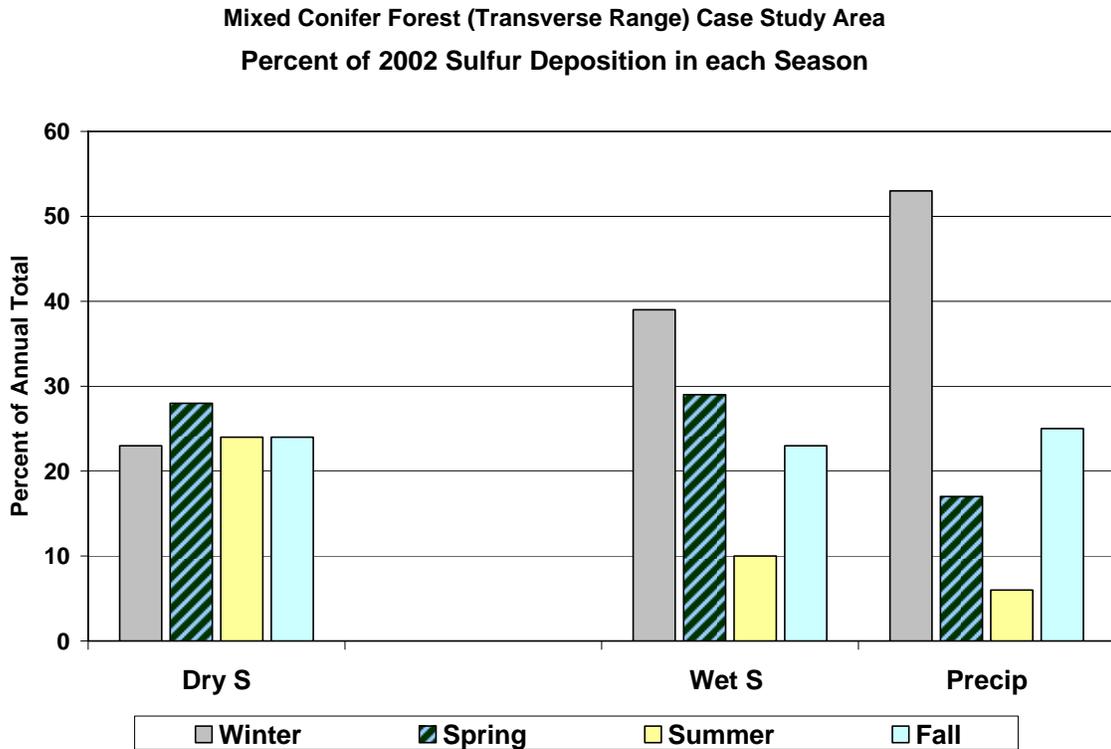


4
5
6

Figure 3.3-15g. Percentage of 2002 deposition for each component of sulfur deposition in the Rocky Mountain National Park.



1
 2 **Figure 3.3-15h.** Percentage of 2002 deposition for each component of sulfur
 3 deposition in the Sierra Nevada Range portion of the Mixed Conifer Forest Case
 4 Study Area.



5
 6 **Figure 3.3-15i.** Percentage of 2002 deposition for each component of sulfur
 7 deposition in the Transverse Range portion of the Mixed Conifer Forest Case
 8 Study Area.

1 **3.3.3.9 Summary of Case Study Analysis Findings**

2 The key findings from the case study analyses are summarized below.

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

5 (2) These regional differences in deposition correspond to the regional differences in NO_x
6 and SO₂ concentrations and emissions, which are also higher in the East.

7 (3) NO_x emissions are much greater and generally more widespread than NH₃ emissions
8 nationwide; high NH₃ emissions tend to be more local (e.g., eastern North Carolina) or sub-
9 regional (e.g., the upper Midwest and Plains States).

10 (4) The relative amounts of oxidized versus reduced nitrogen deposition are consistent
11 with the relative amounts of NO_x and NH₃ emissions.

12 (a) Oxidized nitrogen deposition exceeds reduced nitrogen deposition in most of
13 the case study areas; the major exception being the Neuse River/Neuse River Estuary
14 Case Study Area.

15 (b) Reduced nitrogen deposition exceeds oxidized nitrogen deposition in the
16 vicinity of local sources of NH₃.

17 (5) There can be relatively large spatial variations in both total reactive nitrogen
18 deposition and sulfur deposition within a case study area; this occurs particularly in those areas
19 that contain or are near a high emissions source of NO_x, NH₃, and/or SO₂.

20 (6) The seasonal patterns in deposition differ between the case study areas.

21 (a) For the case study areas in the East, the season with the greatest amounts of
22 total reactive nitrogen deposition correspond to the season with the greatest amount of
23 sulfur deposition. Deposition peaks in spring in the Adirondack, Hubbard Brook
24 Experimental Forest, and Kane Experimental Forest case study areas, and it peaks in
25 summer in the Potomac River/Potomac Estuary, Shenandoah, and Neuse River/Neuse
26 River Estuary case study areas.

27 (b) For the case study areas in the West, there is less consistency in the seasons
28 with greatest total reactive nitrogen and sulfur deposition in a given area. In general, both
29 nitrogen and/or sulfur deposition peaks in spring or summer. The exception to this is the
30 Sierra Nevada Range portion of the Mixed Conifer Forest Case Study Area, in which
31 sulfur deposition is greatest in winter.

3.4 CONTRIBUTIONS OF EMISSIONS OF NO_x AND NH₃ TO DEPOSITION OF NITROGEN

3.4.1 Purpose and Intent

The targeted ecological effect areas' public welfare effects of concern in this review associated with ambient NO_x and SO_x do not occur due to direct exposure to ambient concentrations of NO_x and SO_x, but rather due to deposition of these compounds in the environment. Ecosystem effects occur because of ecological exposures to loadings of all forms of nitrogen and sulfur, and this is due, in part, to atmospheric deposition of nitrogen and sulfur. Atmospheric deposition of nitrogen and sulfur is directly related to the concentrations of NO_x, NH₃, and SO_x in the atmosphere, and thus, reducing atmospheric emissions of NO_x, NH₃, and SO_x will directly impact deposited nitrogen and sulfur and the associated ecosystem effects. In order to set ambient standards for NO_x and SO_x that are protective of public welfare, it is necessary to understand the contribution of ambient NO_x and SO_x to the ecosystem pollutants of concern: sulfur and total reactive nitrogen. Because the focus of this review is on oxides of nitrogen, rather than on total reactive nitrogen, it is important to understand the contribution of NO_x relative to reduced forms of nitrogen (NH₃ and NH₄⁺) to deposition. This section describes the analysis of the contribution of NO_x relative to reduced forms of nitrogen. It also examines the contributions of SO_x emissions to sulfur deposition. These analyses use CMAQ sensitivity runs to estimate the relative percentage contribution of NO_x, NH₃, and SO_x emissions to total nitrogen deposition (the oxidized and reduced forms of nitrogen and total sulfur deposition).

3.4.2 Analytical Techniques

For a more informed understanding of the roles of NO_x, NH₃, and SO_x in deposition of nitrogen and sulfur, the CMAQ model for several sensitivity simulations was run. These simulations include three separate model runs in which anthropogenic emissions of NO_x, NH₃, or SO_x were reduced by 50% from base case emissions levels (i.e., one run for each of the three pollutants). The 2005 12-km CMAQ run for the eastern United States was used as the base case for this analysis. The NO_x, NH₃, and SO_x emissions reductions were applied to the 2005 emissions for all states within the eastern modeling domain²⁷. The 50% NO_x reduction scenario

²⁷ The CMAQ model configuration and modeling domain for these applications are described in Appendix 1 of this report.

1 resulted in a NO_x emissions reduction of ~ 9 MM tons. This amount is more than four times the
2 amount of emissions reduced in the 50% NH₃ scenario (~ 2 MM tons). The 50% SO_x emissions
3 reduction scenario removed ~ 7 MM tons of SO_x from states in the eastern modeling domain.

4 Each sensitivity run was performed for January, April, July, and October 2005, to
5 represent differences in emissions and meteorology in each season of that year. The wet and dry
6 deposition predictions from the CMAQ base case and sensitivity runs were used to calculate the
7 4-month average deposition in each grid cell. The results are used to estimate (1) the relative
8 contribution of emissions of NO_x and NH₃ to deposition of total, reduced, and oxidized nitrogen
9 deposition, and (2) the relative contribution of SO_x emissions to sulfur deposition. The focus is
10 on the percentage contribution in the six case study areas of the East.

11 3.4.3 Results and Findings

12 Contributions of NO_x Emissions to Total Reactive Nitrogen Deposition

13 **Figure 3.4-1** shows the impacts of the 50% NO_x scenario on total reactive nitrogen
14 deposition in the East. In general, a 50% reduction in NO_x had a 30% to 40% impact (i.e.,
15 reduction) on total reactive nitrogen deposition. This includes all or most of the Kane
16 Experimental Forest, Potomac River/Potomac Estuary, and Shenandoah case study areas.
17 Portions of the East where NO_x emissions had the least impact on total reactive nitrogen
18 deposition, including the Neuse River/Neuse River Estuary Case Study Area, generally
19 correspond to areas of highest NH₃ emissions.

20 To further explore the relationships between NO_x emissions and total reactive nitrogen
21 deposition, the impact on oxidized and reduced nitrogen deposition, as shown in **Figures 3.4-2**
22 and **3.4-3**, was examined. These figures reveal that the 50% reduction in NO_x emissions resulted
23 in a 40% to 50% reduction in oxidized nitrogen deposition, indicating that nearly all of the
24 oxidized nitrogen deposition is due to NO_x emissions. The Potomac River/Potomac Estuary,
25 Shenandoah, and Neuse River/Neuse River Estuary case study areas each had reductions in
26 oxidized nitrogen of 45% to 50%. The impacts were somewhat less in the Adirondack, Hubbard
27 Brook Experimental Forest, and Kane Experimental Forest case study areas.

28 The 50% reduction in NO_x generally had a small impact on reduced nitrogen deposition
29 across the East (\pm 6%). Some case study areas had lower reduced nitrogen, whereas others had
30 slight increases. The Adirondack, Kane Experimental Forest, and Hubbard Brook Experimental
31 Forest case study areas all had lower reduced nitrogen deposition. However, in the Neuse

1 River/Neuse River Estuary Case Study Area and in portions of the Potomac River/Potomac
2 Estuary and Shenandoah case study areas, the NO_x emissions impacts are slightly positive,
3 suggesting that NO_x emissions contribute to greater deposition of reduced nitrogen. This
4 relationship reflects the atmospheric reactions that lead to deposition of reduced nitrogen. One
5 possible explanation for this is that reducing NO_x reduces HNO_3 , which limits NO_3^- formation,
6 thereby increasing the lifetime of NH_3 . This may result in a net increase in $\text{NH}_3/\text{NH}_4^+$
7 deposition. Because the deposition velocity of NH_3 is much higher than the deposition velocity
8 for NH_4^+ aerosol, dry deposition of NH_3 increases closer to sources of NH_3 .

9 **Contributions of NH_3 Emissions to Total Reactive Nitrogen Deposition**

10 **Figure 3.4-4** shows the relative impact of emissions of the 50% NH_3 scenario on
11 deposition of total reactive nitrogen. The locations with the greatest contributions from NH_3
12 emissions are generally the same locations where the contribution from NO_x is the least. Portions
13 of the Potomac River/Potomac Estuary, Shenandoah, and Neuse River/Neuse River Estuary case
14 study areas, where there are local emissions of NH_3 , show contributions to total reactive nitrogen
15 deposition approaching 50%. Elsewhere in these case study areas and in the Adirondack, Kane
16 Experimental Forest, and Hubbard Brook Experimental Forest case study areas, the contribution
17 of total reactive nitrogen is generally 10% to 20% or less.

18 **Figures 3.4-5** and **3.4-6** explore the relationship between NH_3 emissions and nitrogen
19 deposition in more detail, examining separately the relative impacts of NH_3 on oxidized and
20 reduced forms of nitrogen. In the Potomac River/Potomac Estuary, Shenandoah, Kane
21 Experimental Forest, and Neuse River/Neuse River Estuary case study areas, the 50% NH_3
22 emissions scenario results in a 40% to 50% impact, indicating that nearly all of the reduced
23 nitrogen in these areas is likely associated with NH_3 emissions. The contributions from NH_3 to
24 reduced nitrogen deposition were somewhat less (generally 30% to 40%) for the Adirondack and
25 Hubbard Brook Experimental Forest case study areas. Also, in the Potomac River/Potomac
26 Estuary, Shenandoah, and Neuse River/Neuse River Estuary case study areas, the NH_3 scenario
27 resulted in a slight increase in oxidized nitrogen deposition. This relationship reflects the
28 atmospheric reactions that lead to the deposition of reduced and oxidized nitrogen. Reducing
29 NH_3 limits NO_3^- aerosol formation, increasing the lifetime of HNO_3 . The ratio of HNO_3 to
30 nitrate (NO_3^-) increases, and because the deposition velocity of HNO_3 is much larger than that of
31 NO_3^- aerosol, dry deposition of total oxidized nitrogen increases. In the Adirondack, Kane

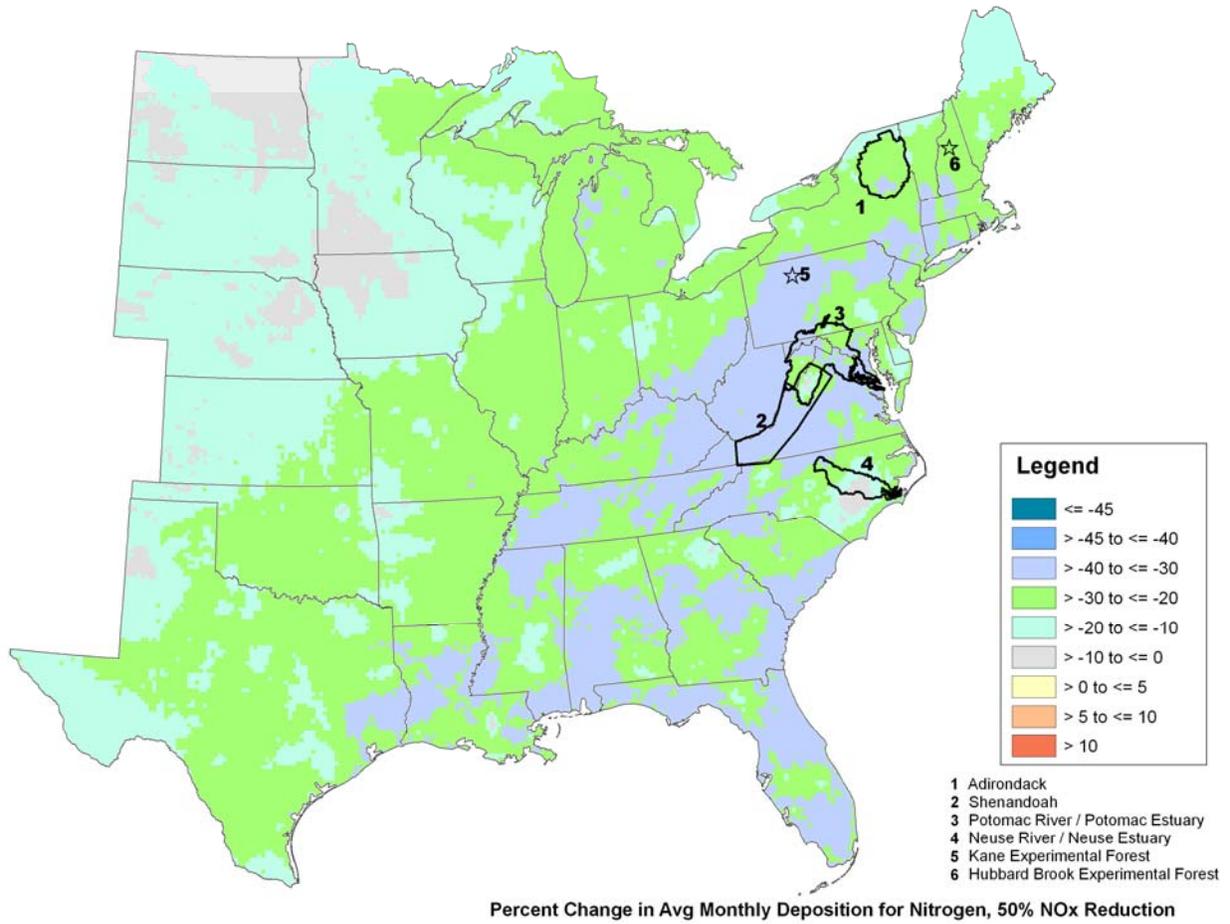
1 Experimental Forest, and Hubbard Brook Experimental Forest case study areas, the 50% NH₃
2 scenario produced a small decrease (up to 2%) in oxidized nitrogen deposition.

3 **Contributions of SO₂ Emissions to Sulfur Deposition**

4 As shown in **Figure 3.4-7**, a 50% reduction in SO_x emissions resulted in nearly a 50%
5 reduction in sulfur deposition in the Kane Experimental Forest, Potomac River/Potomac Estuary,
6 Shenandoah, and Neuse River/Neuse River Estuary case study areas. The contribution is
7 somewhat less in the Adirondack and Hubbard Brook Experimental Forest case study areas,
8 which are more distant from sources of high SO₂ emissions compared with the other case study
9 areas. In general, the contribution of SO₂ emissions to sulfur deposition is fairly linear for the
10 50% reduction scenario that was modeled.

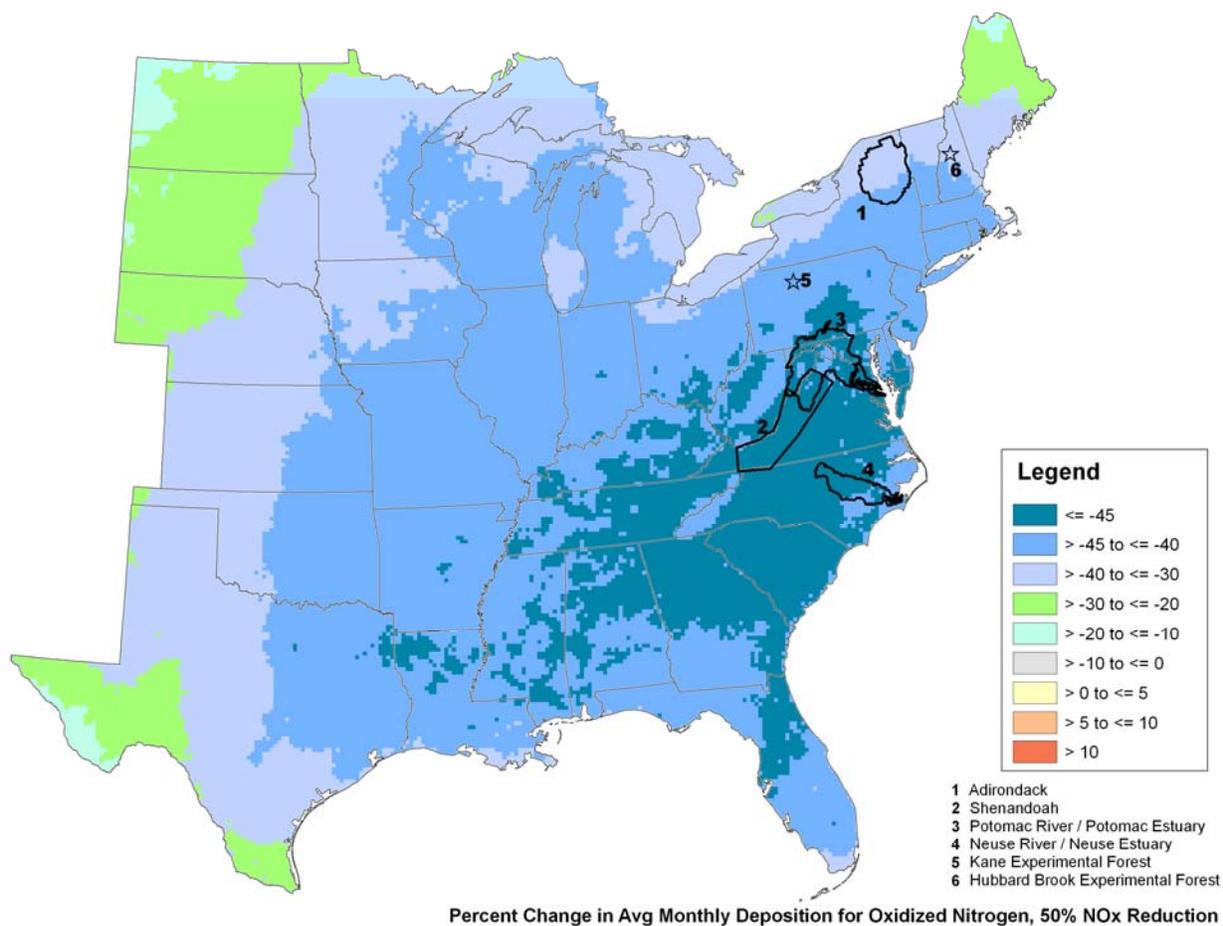
11 **3.4.4 Summary of Findings**

12 From this study of the contribution of emissions to deposition in the East, it is found that
13 NO_x emissions have significant impacts on total nitrogen deposition and account for almost all of
14 the oxidized nitrogen deposition. The contributions of NO_x emissions compared with NH₃
15 emissions appear to be separable in that NO_x affects mainly oxidized nitrogen whereas NH₃
16 affects mainly reduced nitrogen. Because oxidized nitrogen deposition is a greater portion of
17 total reactive nitrogen deposition in most areas, NO_x emissions contribute more to total reactive
18 nitrogen than emissions of NH₃. However, local NH₃ emissions do make significant
19 contributions to total reactive nitrogen deposition near the sources of these emissions.



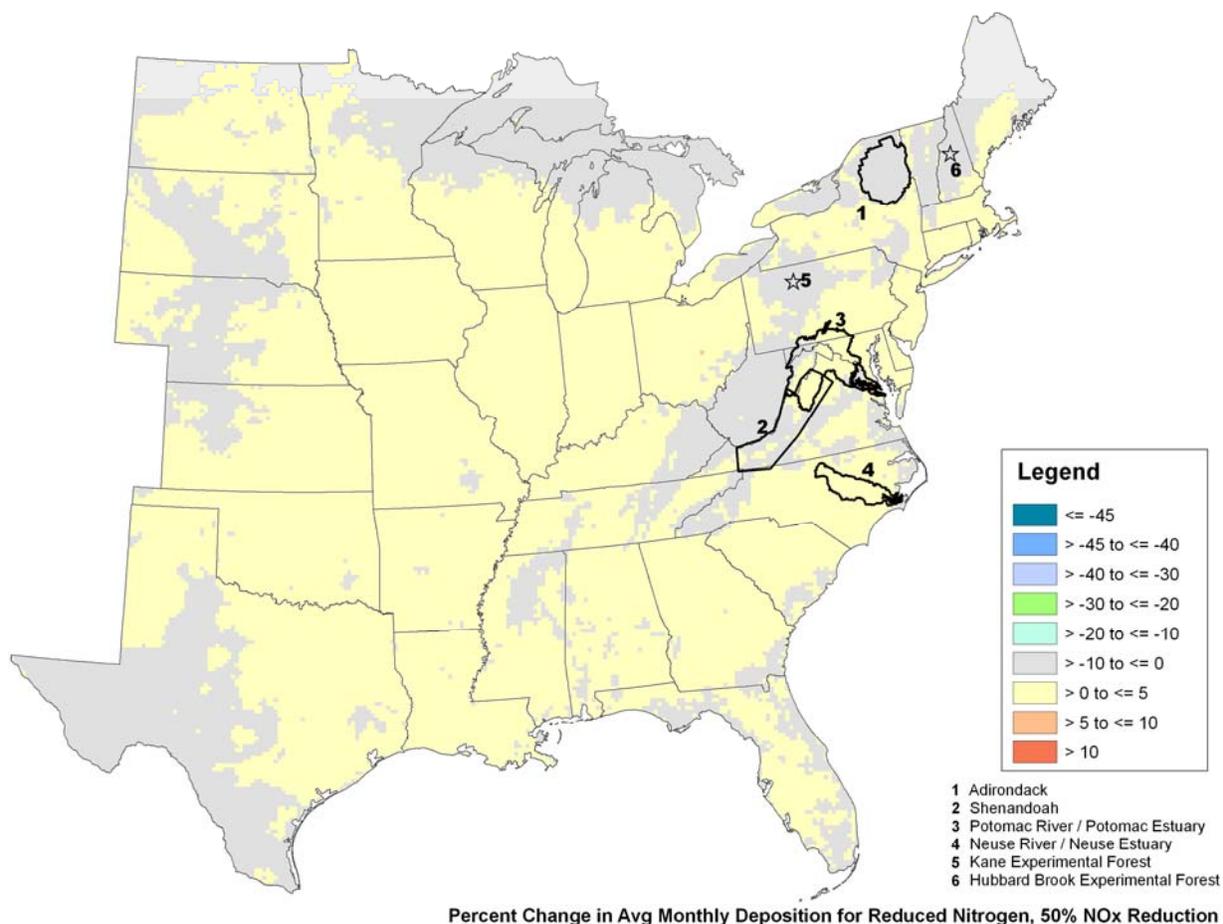
1
2
3

Figure 3.4-1. The percentage impacts of a 50% decrease in NO_x emissions on total reactive nitrogen deposition in the East.



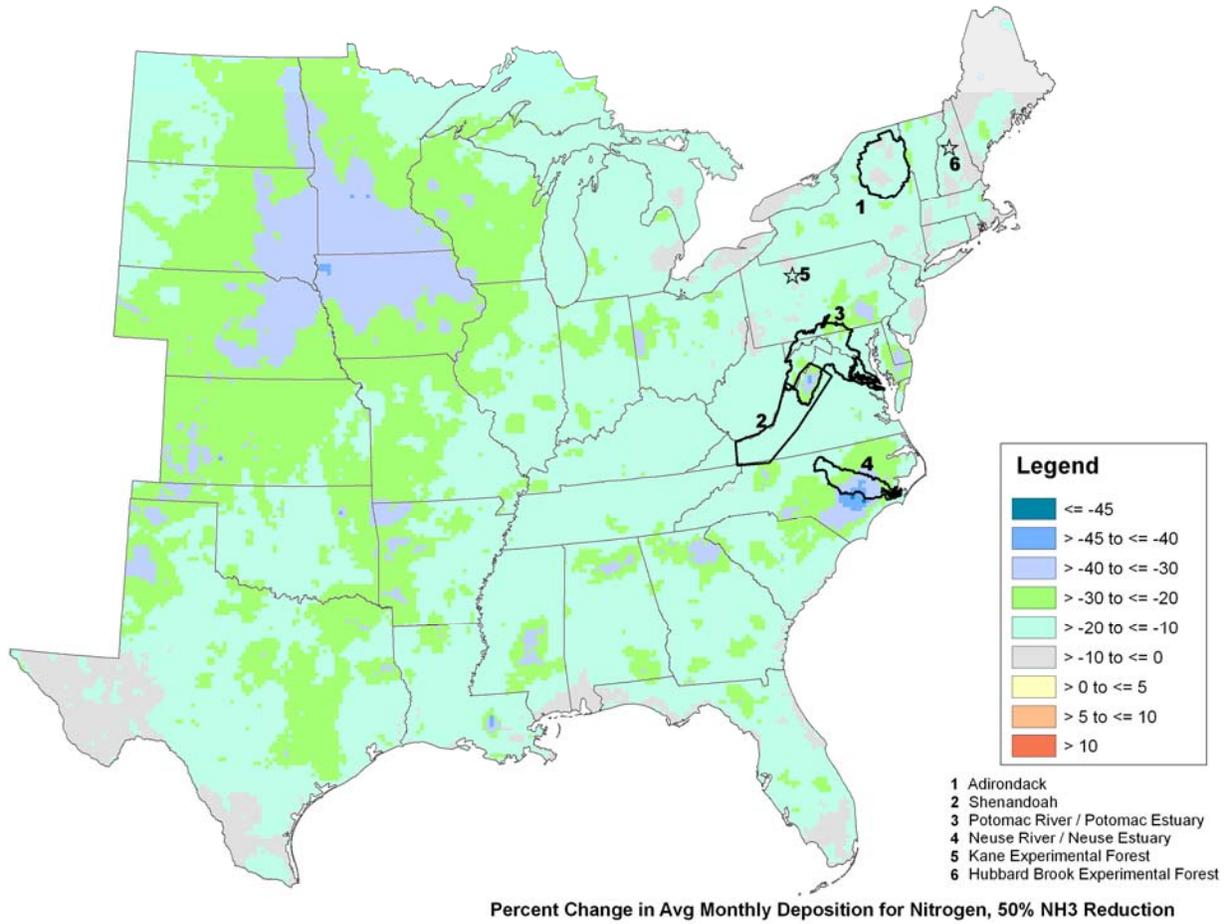
1
2
3

Figure 3.4-2. The percentage impacts of a 50% decrease in NO_x emissions on oxidized nitrogen deposition in the East.



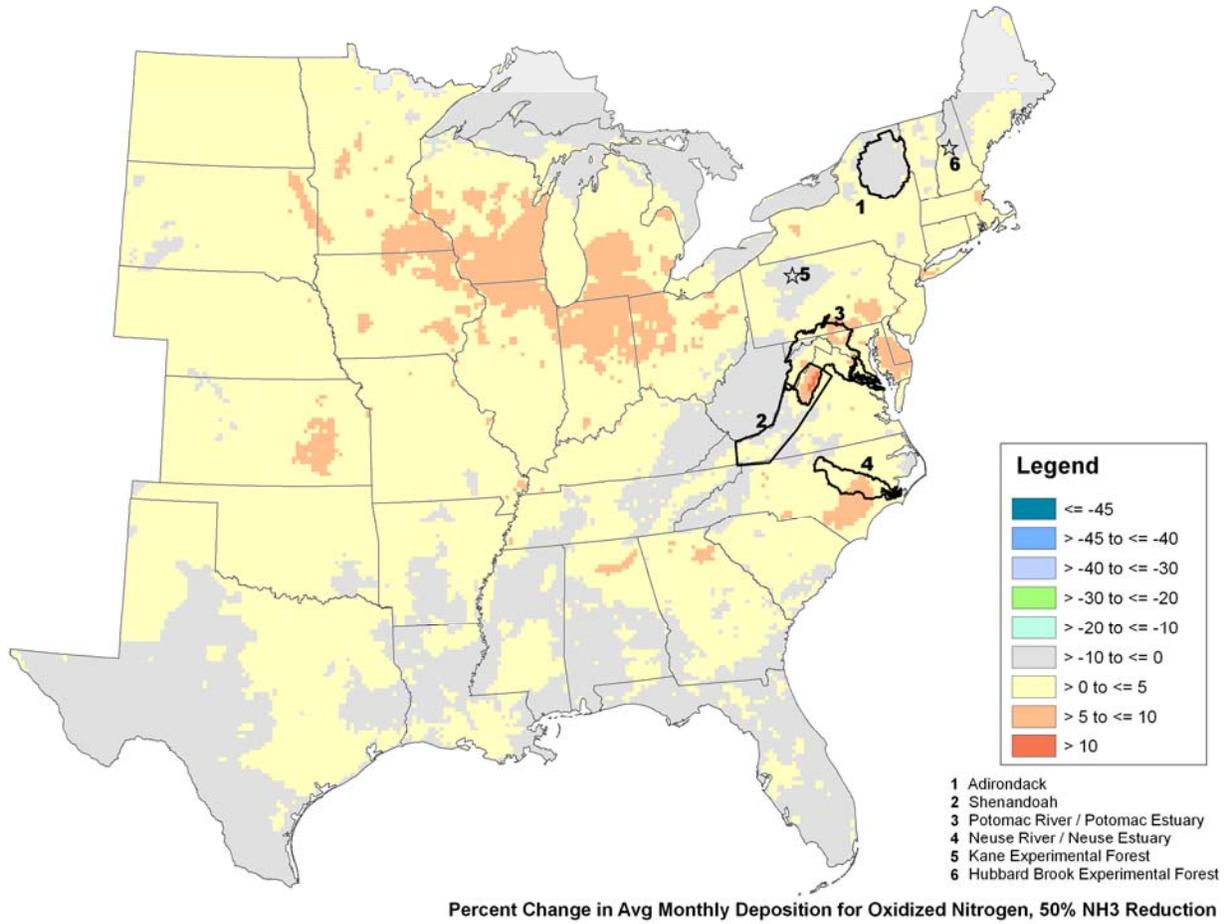
1
2
3

Figure 3.4-3. The percentage impacts of a 50% decrease in NO_x emissions on reduced nitrogen deposition in the East.



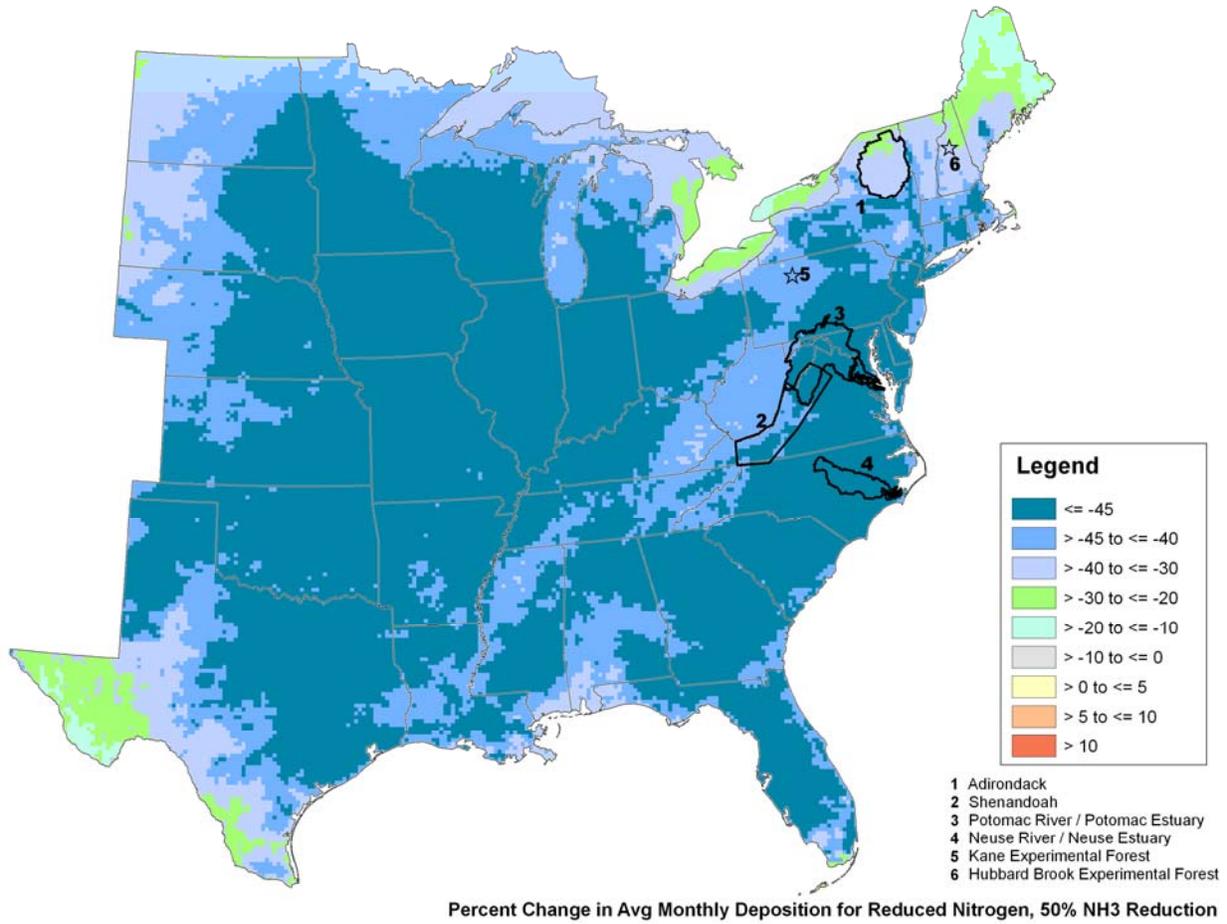
1
2
3

Figure 3.4-4. The percentage impacts of a 50% decrease in NH₃ emissions on total reactive nitrogen deposition in the East.



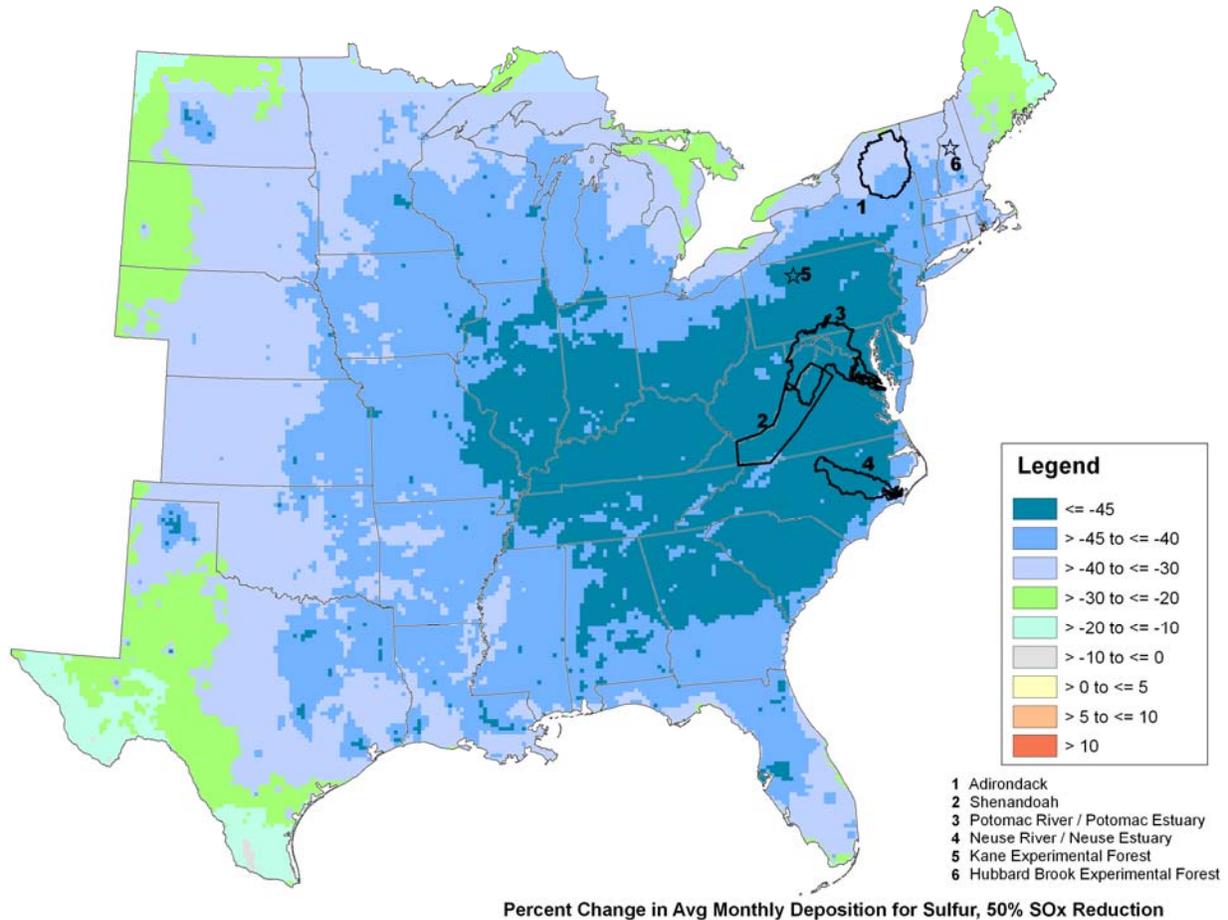
1
2
3

Figure 3.4-5. The percentage impacts of a 50% decrease in NH₃ emissions on oxidized nitrogen deposition in the East.



1
2
3

Figure 3.4-6. The percentage impacts of a 50% decrease in NH₃ emissions on reduced nitrogen deposition in the East.



1

2

3

Figure 3.4-7. The percentage impacts of a 50% decrease in SO_x emissions on sulfur deposition in the East.

4

3.5 DISCUSSION OF UNCERTAINTIES

5

This chapter provides a nationwide overview of NO_x, SO_x, and NH₃ emissions; NO_x and

6

SO_x concentrations; and nitrogen and sulfur deposition, as well as, a more focused

7

characterization of nitrogen and sulfur deposition for the aquatic and terrestrial case study areas.

8

These analyses are based on measured data and model predictions that each contain a number of

9

areas of uncertainty. This section identifies and describes uncertainties associated with the

10

various aspects of this analysis, but does not to quantify these uncertainties.

11

The uncertainties associated with emissions data of NO_x and SO_x will vary based on the

12

method used to determine or estimate emissions. The smallest uncertainties are likely to be

13

associated with EGUs, whose emissions are determined by continuous emissions monitoring. For

14

many other source categories, emissions are based the application of emissions factors to the

1 sector's activity data. Uncertainties in emissions may increase for a particular source category if
2 the types and extent of source measurements and analytical procedures used to derive emissions
3 factors are not fully representative of the source category for which they are applied. For some
4 source categories, the calculations of emissions involve complex models that may not fully
5 represent actual levels of emissions in a particular location at a particular time. In addition,
6 activity data used in "top-down" inventories which allocate national emissions to individual
7 counties may not properly reflect local emissions for all areas.

8 Areas of uncertainty in characterizing NO_x and SO_x concentrations and nitrogen and
9 sulfur deposition levels include uncertainties in monitoring instrumentation and measurement
10 protocols, as well as limitations in the spatial extent of existing monitoring networks for these
11 pollutant species. In addition, as described elsewhere in this chapter, there are no "true"
12 measurements of dry deposition. In view of geographic limitations in monitoring activities for
13 some species, predictions of CMAQ are relied upon to characterize NO_x and SO_x concentrations
14 and nitrogen and sulfur dry deposition. Although CMAQ is a "state-of-the-science"
15 photochemical model, uncertainties in CMAQ, like those in other photochemical models, arise
16 due to uncertainties in model formulation and in the inputs which drive the simulation chemistry
17 and transport processes within the model. The model formulation uncertainties most relevant for
18 this assessment include the aspects of the non-linear photochemical processes that determine the
19 chemical form and transformations of NO_x and SO_x in the atmosphere over multiday time
20 periods and of the processes that affect the removal of NO_x and SO_x through deposition. In
21 addition to the emissions uncertainties identified above, a key uncertainty in the input emissions
22 estimates may be the extent of re-emissions of NH₃ (i.e., "bi-directional flux") that is not
23 accounted for in air quality models and would affect ammonia and ammonium deposition and the
24 neutralization of sulfuric acid and nitric acid in the formation sulfate and nitrate particles,
25 respectively. Uncertainties in meteorological inputs including the presence of clouds, the
26 occurrence and amount of precipitation, and the extent of vertical mixing affect the uncertainty
27 in model predictions of pollutant concentrations and deposition. The degree of uncertainty in
28 these inputs may be greater in complex terrain, which is an important factor for those sensitive
29 ecosystems located in mountainous areas.

30 A model performance evaluation of CMAQ-predicted concentrations and deposition was
31 conducted using measurements at CASTNET and NADP sites, respectively. The results of this

1 evaluation are provided in Appendix 1. The purpose of this evaluation is to determine the degree
2 of comparability between predictions and observations to provide confidence in the use of
3 CMAQ for this assessment. The model performance statistics do not necessarily represent a
4 quantitative estimate of model uncertainty since, aside from uncertainties in the modeling
5 system, uncertainties exist in the measurements and uncertainty is introduced by the
6 incommensurability between the grid cell average model predictions and the point measurements
7 at monitoring sites.

8 Another aspect of uncertainty applicable to this analysis is associated with the
9 combination of wet deposition from NADP measurements with dry deposition from CMAQ. For
10 example, uncertainties in the modeling system may result in times when the transport patterns
11 and precipitation events simulated in the model do not fully align in space and time with actual
12 atmospheric conditions in a particular location. This may result physical and chemical
13 inconsistencies between the measured wet deposition and the modeled dry deposition.

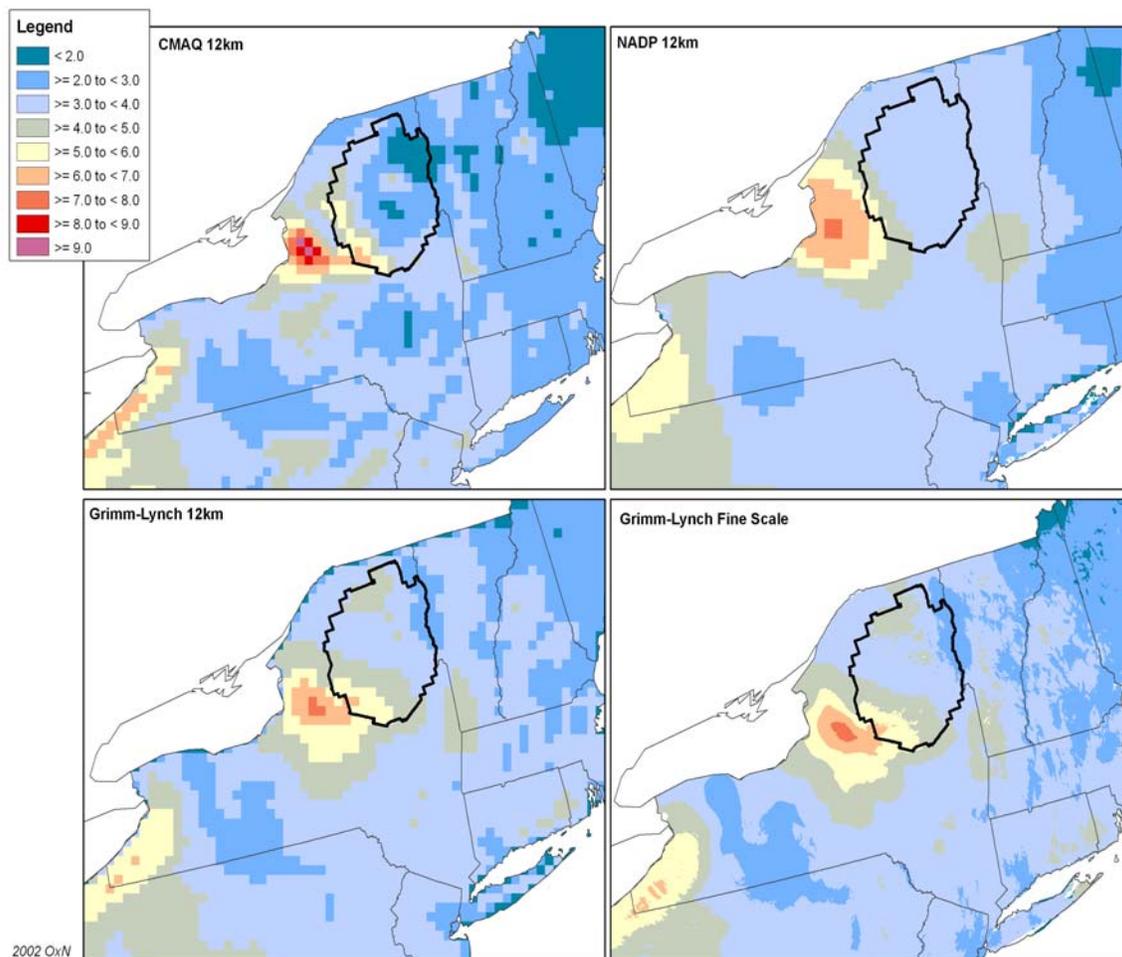
14 In addition, uncertainties are associated with the spatial resolutions of the measured and
15 modeled data used in this analysis. This includes uncertainties associated with (1) gridding the
16 NADP measurements to a 12-km resolution and (2) the representativeness of 12-km data for
17 characterizing deposition in the case study areas, especially for those areas with complex terrain.
18 To examine the latter issue, the 2002 12-km gridded NADP deposition fields were compared to
19 outputs from a high-resolution wet deposition model²⁸ (Grimm-Lynch) which provides fine scale
20 estimates of deposition for 2002 based on an integration of measured precipitation and wet
21 deposition and topography. The CMAQ 12-km gridded wet deposition predictions were also
22 included in this comparison since these data were used in section 3.3.3.4 to characterize seasonal
23 trends in deposition. For the purposes of this analysis, the Grimm-Lynch data was used as the
24 benchmark even though there are also uncertainties in this data set.

25 The analysis of spatial resolution was conducted for the Adirondack Case Study Area
26 because this area has the highest elevations and the most complex terrain of all the case study
27 areas in the eastern United States. The comparison of gridded data includes annual wet
28 deposition of oxidized and reduced nitrogen and sulfur for 2002 for (a) 12-km CMAQ data, (b)
29 12-km NADP data, (c) fine-scale Grimm-Lynch data, and (d) an aggregation of the fine-scale
30 data to 12 km. The 12-km aggregation of the fine-scale data was included to isolate the effects of

²⁸ Grimm, J.W. and J.A. Lynch. Enhanced Wet Deposition Estimates Using Modeled Precipitation Inputs.
Environmental Monitoring and Assessment 90: 243-268, 2004.

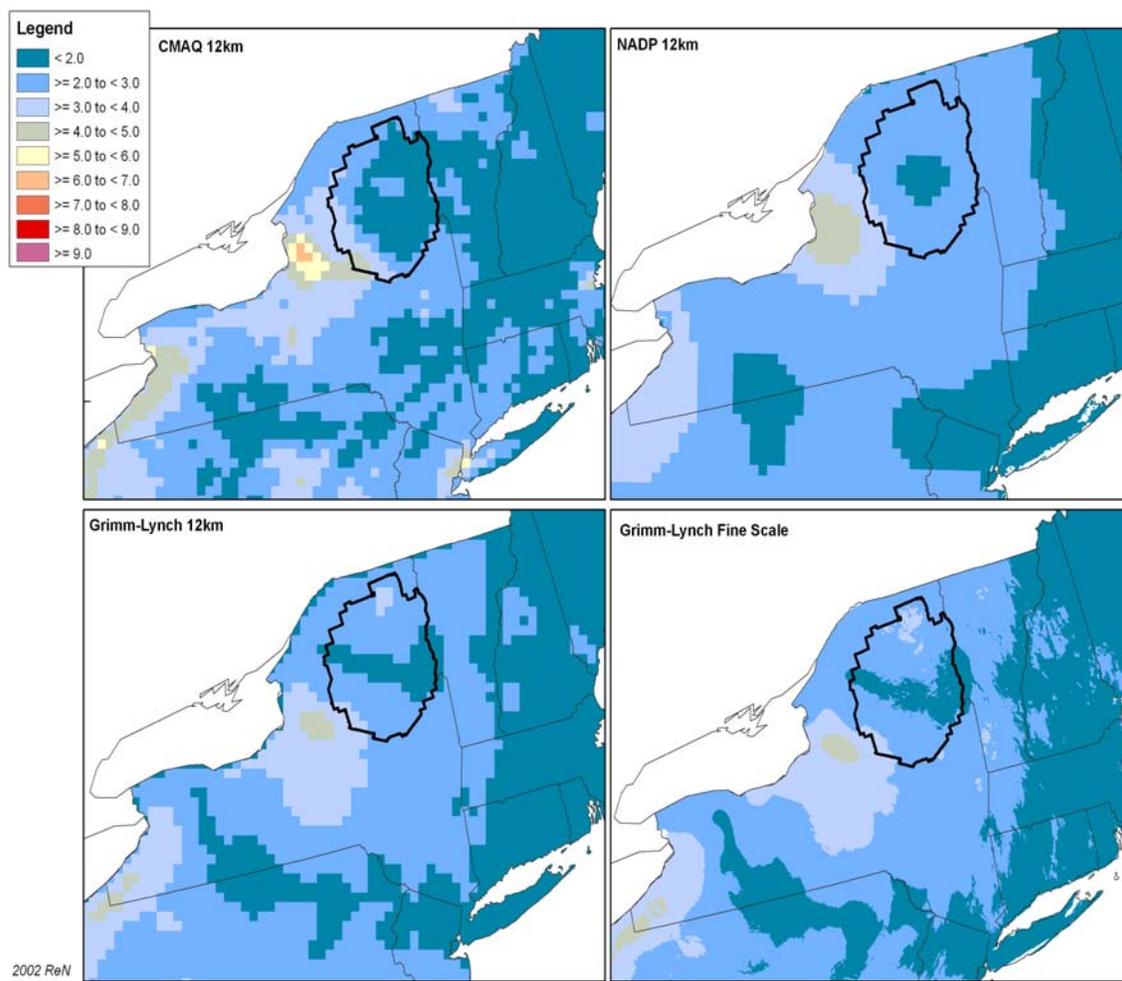
1 grid resolution from the confounding effects introduced by other properties and uncertainties of
2 the CMAQ and NADP data sets. Maps showing the magnitude and spatial patterns of wet
3 deposition for the four data sets are provided in Figures 3.5-1, 3.5-2, and 3.5-3 for oxidized and
4 reduced nitrogen deposition and for sulfur deposition, respectively. The figures reveal both
5 similarities in differences in wet deposition. Comparing the native fine-scale Grimm-Lynch data
6 to the 12-km aggregate of these data indicates only slight, very local differences between the
7 fine-scale and 12-km deposition for each of the three deposition species. Thus, it does not appear
8 that the use of the 12-km resolution data masks any significant terrain-induced features of
9 deposition, at least for this case study area. There are both similarities and notable differences
10 between the CMAQ, NADP, and Grimm-Lynch deposition fields at 12 km. Again, using the
11 Grimm-Lynch predictions as the benchmark, the NADP fields are perhaps too smooth while the
12 CMAQ predictions tend to show enhanced spatial gradients. All three data sets show an area of
13 relatively high wet deposition which extends westward from Lake Ontario across the southwest
14 portion of the Adirondack Case Study Area. The Grimm-Lynch data also suggest that a
15 secondary maximum of wet deposition extends from the northern border of the Adirondack Case
16 Study Area southward into the central portion of the area. The CMAQ shows this feature as a
17 small area of high deposition near the central part of the Adirondack Case Study Area. The
18 secondary maximum does not appear to be captured by the NADP 12-km gridded data. Overall,
19 the spatial patterns in nitrogen and sulfur deposition across the Adirondacks seen from the three
20 data sets examined here are similar to the patterns in NO_3^- and SO_4^{2-} wet deposition ,
21 respectively, found by Ito, Mitchell, and Driscoll (2002) based on an analysis of measured
22 precipitation, temperature, precipitation chemistry, elevation and other factors.

23 Although there are uncertainties in the data, models, and techniques used for this
24 assessment, this analysis relies upon the most applicable measurements and state-of-the-science
25 models. In addition, these data and models are used in a manner that considers their relative
26 strengths and limitations. The inherent uncertainties are not expected to measurably affect the
27 robustness of these conclusions and findings on the characterization of concentrations and
28 deposition.

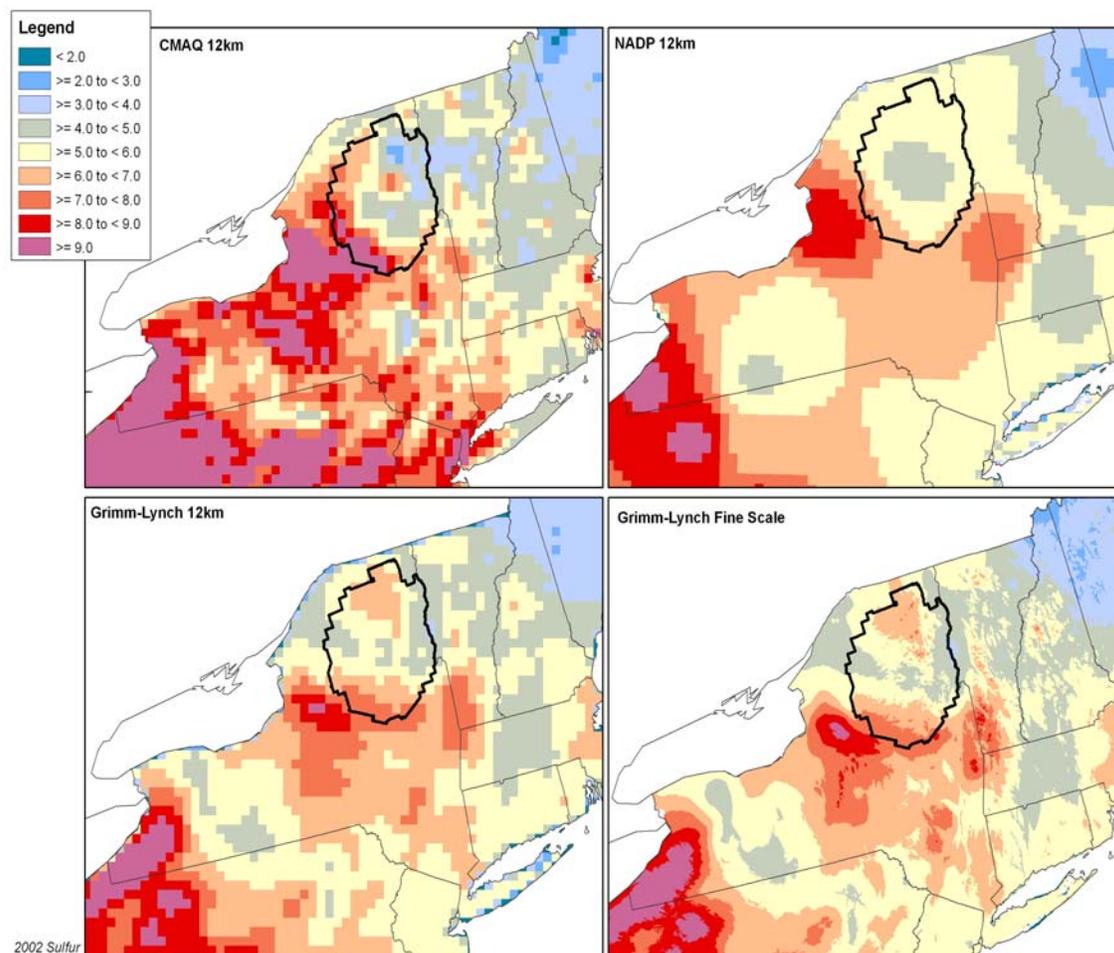


1
2
3

Figure 3.5-1. Fine-scale and 12-km annual total oxidized nitrogen deposition for the Adirondack Case Study Area and the surrounding region.



1
2 **Figure 3.5-2.** Fine-scale and 12-km annual total reduced nitrogen deposition for
3 the Adirondack Case Study Area and the surrounding region.



1
2
3
4
5
6
7
8
9
10
11

Figure 3.5-3. Fine-scale and 12-km annual total sulfur deposition for the Adirondack Case Study Area and the surrounding region.

3.6 REFERENCES

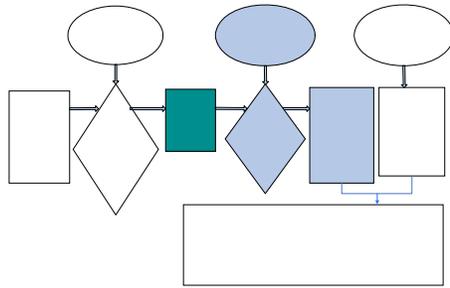
Byun, D.W., and K.L. Schere. 2006. Review of the governing equations, computational algorithms, and other components of the models-3 Community Multiscale Air Quality (CMAQ) modeling system. *Journal of Applied Mechanics Reviews* 59(2):51–77.

Clarke, J.F., E.S. Edgerton, and B.E. Martin. 1997. Dry deposition calculations for the Clean Air Status and Trends Network. *Atmospheric Environment* 31:3667–3678.

Holland, H.D. 1978. *The Chemistry of the Atmosphere and Oceans*. New York: John Wiley & Sons.

- 1 Ito, M. Mitchell, M.J. and Driscoll, C.T. 2002. Spatial patterns of precipitation quantity and
2 chemistry and air temperature in the Adirondack region of New York. *Atmospheric*
3 *Environment Vol. 36*: 1051-1062
- 4 Levine, J.S., T. Bobbe, N. Ray, R.G. Witt, and A. Singh. 1999. *Wildland Fires and the*
5 *Environment: A Global Synthesis*. Report no. UNEP/DEIAEW/TR.99.1. United Nations
6 Environment Programme (UNEP), Division of Environmental Information, Assessment
7 and Early Warning (DEIA&EW), Nairobi, Kenya. Available at
8 <http://www.na.unep.net/publications/wildfire.pdf>.
- 9 Seinfeld, J.H., and S.N. Pandis. 1998. *Atmospheric Chemistry and Physics: From Air Pollution*
10 *to Climate Change*. New York: John Wiley-Interscience Publishers.
- 11 Sickles, J.E., and D.S. Shadwick. 2007a. Changes in air quality and atmospheric deposition in
12 the eastern United States: 1990-2004. *Journal of Geophysical Research 112*(D17).
- 13 Sickles, J.E., II, and Shadwick, D.S. 2007b. Seasonal and regional air quality and atmospheric
14 deposition in the eastern United States, *Journal of Geophysical Research, Vol. 112*:
15 D17302.
- 16 U.S. EPA (Environmental Protection Agency). 1999. *Science Algorithms of EPA Models-3*
17 *Community Multiscale Air Quality (CMAQ) Modeling System*. D.W. Byun and J.K.S.
18 Ching, eds. EPA/600/R-99/030. U.S. Environmental Protection Agency, National
19 Exposure Research Laboratory, Research Triangle Park, NC.
- 20 U.S. EPA (Environmental Protection Agency). 2006. *2002 National Emissions Inventory Data &*
21 *Documentation*. U.S. Environmental Protection Agency, Technology Transfer Network,
22 Clearinghouse for Inventories & Emissions Factors, Office of Air Quality Planning and
23 Standards, Research Triangle Park, NC. Available at
24 <http://www.epa.gov/ttn/chief/net/2002inventory.html> (accessed May 28, 2009).
- 25 U.S. EPA (Environmental Protection Agency). 2008a. *Clean Air Status and Trends Network,*
26 *2007 Annual Report*. U.S. Environmental Protection Agency, Office of Air and

- 1 Radiation, Clean Air Markets Division, Washington, DC. December. Available at
2 http://www.epa.gov/castnet/docs/annual_report_2007.pdf.
- 3 U.S. EPA (Environmental Protection Agency). 2008b. *Integrated Science Assessment (ISA) for*
4 *Oxides of Nitrogen and Sulfur–Ecological Criteria (Final Report)*. EPA/600/R-
5 08/082F. U.S. Environmental Protection Agency, National Center for Environmental
6 Assessment–RTP Division, Office of Research and Development, Research Triangle
7 Park, NC. Available at <http://cfpub.epa.gov/ncea/cfm/recordisplay.cfm?deid=201485>.
- 8 U.S. EPA (Environmental Protection Agency). 2009. *Clean Air Status and Trends Network*
9 *(CASTNET)*. Online information. U.S. Environmental Protection Agency, Office of Air
10 and Radiation, Clean Air Markets Division, Washington, DC. Available at
11 <http://www.epa.gov/CASTNET> (accessed May 2009).
- 12

1
2

3

4.0 ACIDIFICATION

4.1 SCIENCE OVERVIEW

7 Air emissions of sulfur oxides (SO_x), nitrogen oxides
 9 (NO_x), and reduced forms of nitrogen (NH_x) react in the
 11 atmosphere through a complex mix of reactions and
 13 thermodynamic processes in gaseous, liquid, and solid
 14 phases to form various acidifying compounds. These compounds are removed from the
 15 atmosphere through wet (e.g., rain, snow), occult (e.g., fog, mist), or dry (e.g., gases, particles)
 16 deposition. Deposition of SO_x , NO_x , and NH_x leads to ecosystem exposure to acidification. The
 17 *Integrated Science Assessment (ISA) for Oxides of Nitrogen and Sulfur–Ecological Criteria*
 18 *(Final Report)* (ISA) (U.S. EPA, 2008) reports that acidifying deposition has altered major
 19 biogeochemical processes in the United States by increasing the sulfur and nitrogen content of
 20 soils, accelerating sulfate (SO_4^{2-}) and nitrate (NO_3^-) leaching from soil to drainage water,
 21 depleting base cations (especially calcium [Ca^{2+}] and magnesium [Mg^{2+}]) from soils, and
 22 increasing the mobility of aluminum (Al) (U.S. EPA, 2008, Section 3.2.1)

Acidification is the decrease of **acid neutralizing capacity** in water or **base saturation** in soil caused by natural or anthropogenic processes.

23 The extent of soil acidification is a critical factor that regulates virtually all acidification-
 24 related ecosystem effects from sulfur and nitrogen deposition. Soil acidification occurs in
 25 response to both natural factors and acidifying deposition (U.S. EPA, 2008, Section 3.2.1).
 26 Under natural conditions (i.e., low atmospheric deposition of nitrogen and sulfur), the limited
 27 mobility of anions in the soil controls the rate of base cation leaching. However, acidifying
 28 deposition of nitrogen and sulfur species can significantly increase the concentration of anions in
 29 the soil, leading to an accelerated rate of base cation leaching, particularly the leaching of Ca^{2+}
 30 and Mg^{2+} cations.

1 Acidification can impact the health of terrestrial and aquatic ecosystems. One of the
2 effects of soil acidification is the increased mobility of inorganic Al, which is toxic to tree roots,
3 fish, algae, and aquatic invertebrates (U.S. EPA, 2008, Sections 3.2.1.5, 3.2.2.1, and 3.2.3).

4 Both the aquatic and terrestrial effects of acidification have been studied and are
5 highlighted in this chapter. For each effect, information is presented on the following:

- 6 ▪ Ecological indicators, ecological responses, and ecosystem services
- 7 ▪ Characteristics of areas sensitive to acidification
- 8 ▪ Case studies' selection
- 9 ▪ Current conditions in case study areas
- 10 ▪ The ability to extrapolate case study findings to larger areas
- 11 ▪ Current conditions for other areas
- 12 ▪ Ecological effect functions
- 13 ▪ Uncertainty and variability identified for the case studies.

14 The case studies on aquatic acidification and terrestrial acidification were performed as
15 part of this Risk and Exposure Assessment (Appendices 4 and 5, respectively) to aid in
16 determining whether a link can be established between NO_x and SO_x deposition and ecosystem
17 response. These case studies are also intended to test whether area-based risk and exposure
18 assessments are a suitable method for predicting acidification effects on other ecosystems and
19 geographic regions. The studies facilitate extrapolation of impacts from smaller-scale (yet
20 representative) areas to other sensitive areas in the country.

21 **4.1.1 Aquatic Acidification**

22 The changes in major biogeochemical processes and soil conditions caused by acidifying
23 deposition have significant ramifications for the water chemistry and biological functioning of
24 associated surface waters. Surface water chemistry indicates the negative effects of acidification
25 on the biotic integrity of freshwater ecosystems. Because surface water chemistry integrates the
26 sum of soil and water processes that occur upstream within a watershed, it also reflects the
27 results of watershed-scale terrestrial effects, including nitrogen saturation, forest decline, and soil
28 acidification (Stoddard et al., 2003). Thus, water chemistry integrates and reflects changes in soil
29 and vegetative properties and biogeochemical processes (U.S. EPA, 2008, Section 3.2.3.1).

The Aquatic Acidification Case Study, reported in Appendix 4 and summarized in this chapter, is intended to estimate the ecological exposure and risk posed to aquatic ecosystems from the acidification effects of the deposition of nitrogen and sulfur for two sensitive regions of the eastern United States: the Adirondack Mountains and Shenandoah National Park (Virginia) and the surrounding areas of Virginia (henceforth referred to as the Adirondack Case Study Area and the Shenandoah Case Study Area, respectively).

4.1.2 Terrestrial Acidification

Deposition of NO_x and SO_x can result in acidification of certain terrestrial ecosystems. Terrestrial acidification occurs as a result of both natural biogeochemical processes and acidifying deposition where strong acids are deposited into the soil. If soil base saturation (i.e., the concentration of exchangeable base cations as a percentage of the total cation exchange capacity, or the sum total of exchangeable cations that a soil can absorb) is 20% to 25%, or lower, inorganic Al can become mobilized, leading to the leaching of Al into soil waters and surface waters (Reuss and Johnson, 1985). Because ecosystems and species may respond differently, case studies have been used to illustrate the potential effects of acidification on sensitive species. Section 4.3 of this chapter presents the quantitative approach used to analyze the acidification effects of total nitrogen, NO_x (as a component of total nitrogen), and SO_x deposition on red spruce and sugar maple.

4.2 AQUATIC ACIDIFICATION

When sulfur or nitrogen migrates 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 the balance of electric charge. If the countercharge is provided by base cations, such as calcium (Ca^{2+}), magnesium (Mg^{2+}), sodium (Na^+), or potassium (K^+), rather than hydrogen (H^+) and aluminum (Al^{3+}), the acidity of the soil water is neutralized, but the base saturation of the soil is

For the purpose of this case study, acid neutralizing capacity (ANC) of surface waters is simply measured as the total amount of strong base ions minus the total amount of strong acid anions:

$$\text{ANC} = (\text{Ca}^{2+} + \text{Mg}^{2+} + \text{K}^+ + \text{Na}^+ + \text{NH}_4) - (\text{SO}_4^{2-} + \text{NO}_3^- + \text{Cl}^-)$$

The unit of ANC is usually microequivalents per liter ($\mu\text{eq/L}$). If the sum of the equivalent concentrations of the base cations exceeds those of the strong acid anions, then the ANC of a waterbody will be positive. To the extent that the base cation sum exceeds the strong acid anion sum, the ANC will be higher. Higher ANC is generally associated with high pH and Ca^{2+} concentrations; lower ANC is generally associated with low pH and Al^{3+} concentrations and a greater likelihood of toxicity to biota.

1 reduced. Continued SO_4^{2-} or NO_3^- leaching can further deplete the base cation supply of the soil.
 2 As the base cations are removed, continued deposition and leaching of SO_4^{2-} and/or NO_3^- (with
 3 H^+ and Al^{3+}) leads to acidification of soil water, and by connection, surface water. Loss of soil
 4 base saturation is a cumulative effect that increases the sensitivity of the watershed to further
 5 acidifying deposition.

6 It is important to note that these chemical changes can occur over both long- and short-
 7 term timescales. Short-term (i.e., hours or days) episodic changes in water chemistry have
 8 perhaps the most significant biological effects. Episodic chemistry refers to conditions during
 9 rainstorms or snowmelt when proportionately more drainage water is routed through upper soil
 10 horizons, which tends to provide less neutralizing of atmospheric acidity as compared with
 11 deeper soil horizons. Surface water chemistry has lower pH and acid neutralizing capacity
 12 (ANC) during storm runoff or snowmelt than during baseflow conditions. One of the most
 13 important effects of acidifying deposition on surface water chemistry is the short-term change in
 14 chemistry that is termed “episodic acidification.” Some streams may have chronic or average
 15 chemistry that is suitable for aquatic biota, but may be subject to occasional episodic
 16 acidification, with lethal consequences. Episodic declines in pH and ANC are nearly ubiquitous
 17 in drainage waters throughout the eastern United States and are caused partly by acidifying
 19 deposition and partly by natural processes.

21 The ISA concludes the following:

- 23 ■ The evidence is sufficient to infer a
 25 causal relationship between acidifying
 27 deposition and changes in
 29 biogeochemistry related to aquatic
 31 ecosystems. The strongest evidence
 33 comes from studies of changes in
 35 surface water chemistry, including
 37 concentrations of SO_4^{2-} , NO_3^- ,
 39 inorganic Al and Ca, surface water pH,
 41 sum of base cations, ANC, and base
 43 cation surplus.
- 45 ■ The evidence is sufficient to infer a

Documented Evidence of Changes in Aquatic Biota Due to Acidifying Deposition

Species

- Mayflies, crustaceans, and mollusks from some streams
- Salmonid fish, smallmouth bass (*Micropterus dolomieu*)
- young-of-the year brook trout.

Community

- Species richness of plankton, invertebrates, and fish
- Invertebrate taxa, including mayflies, amphipods, snails, and clams
- Loss of species diversity and absence of several sensitive fish species
- Early life stages more sensitive to acidic conditions than the young-of-the-year, yearlings, and adults.

(U.S. EPA, 2008, Section 3.2.3.4)

1 causal relationship between acidifying deposition and changes in aquatic biota. The
2 strongest evidence comes from studies of aquatic systems exposed to elevated levels of
3 acidifying deposition that support fewer species of fish, macroinvertebrates, and diatoms.
4 Decreases in ANC and pH and increases in inorganic Al concentration contribute to
5 declines in taxonomic richness of zooplankton, macroinvertebrates, and fish.

6 **4.2.1 Ecological Indicators, Ecological Responses, and Ecosystem Services**

7 **4.2.1.1 Ecological Indicators**

8 Surface water chemistry is a primary indicator of acidification and the resulting negative
9 effects on the biotic integrity of freshwater ecosystems. Chemical receptors can be used to assess
10 effects of acidifying deposition on lake or stream acid-base chemistry. These receptors include
11 surface water pH and concentrations of SO_4^{2-} , NO_3^- , Al, and Ca^{2+} ; the sum of base cations; and
12 the recently developed base cation surplus. Another widely used water chemistry indicator for
13 both atmospheric deposition sensitivity and effects is ANC. Although ANC does not relate
14 directly to the health of biota, the utility of the ANC criterion lies in the association between
15 ANC and the surface water constituents that directly contribute to or ameliorate acidity-related
16 stress, in particular pH, Ca^{2+} , and Al. ANC is also used because it integrates overall acid status
17 and because surface water acidification models do a better job projecting ANC than do pH and
18 inorganic Al concentrations. The Aquatic Acidification Case Study, therefore, used ANC as the
19 indicator of aquatic acidification.

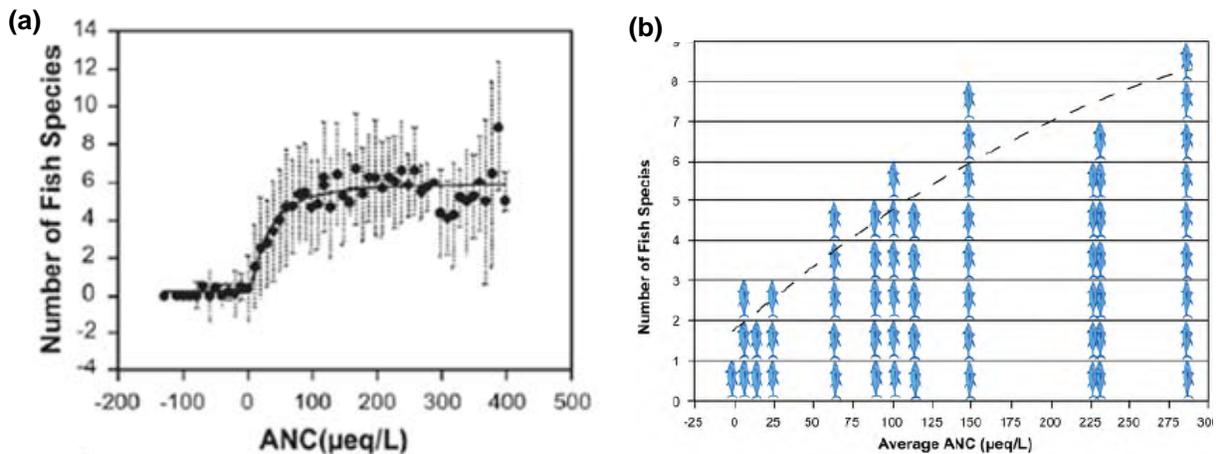
20 Process-based models, such as the Model of Acidification of Groundwater in Catchment
21 (MAGIC) and PnET-BGC (an integrated biogeochemical model), use the ANC calculated from
22 the charge balance.

23 **4.2.1.2 Ecological Responses**

24 Low ANC concentrations have direct effects on aquatic systems (e.g., individual species
25 fitness loss or death, reduced species richness, altered community structure). At the community
26 level, species richness is positively correlated with pH and ANC (Kretser et al., 1989; Rago and
27 Wiener, 1986) because energy cost in maintaining physiological homeostasis, growth, and
28 reproduction is high at low ANC levels (Schreck, 1981, 1982; Wedemeyer et al., 1990). For
29 example, Sullivan et al. (2006) found a logistic relationship between fish species richness and
30 ANC class for Adirondack Case Study Area lakes (**Figure 4.2-1, a**), which indicates the

1 probability of occurrence of an organism for a given value of ANC. In the Shenandoah Case
 2 Study Area, a statistically robust relationship between acid-base status of streams and fish
 3 species richness was also documented (**Figure 4.2-1, b**). In fact, ANC has been found in studies
 4 to be the best single indicator of the biological response and health of aquatic communities in
 5 acid-sensitive systems (Lien et al., 1992; Sullivan et al., 2006).

6 Biota are generally not harmed when ANC values are >100 microequivalents per liter
 7 ($\mu\text{eq/L}$). The number of fish species also peaks at ANC values >100 $\mu\text{eq/L}$ (Bulger et al., 1999;
 8 Driscoll et al., 2001; Kretser et al., 1989; Sullivan et al., 2006). Below 100 $\mu\text{eq/L}$, it has been
 9 shown that fish fitness and community diversity begin to decline (**Figure 4.2-1**). At ANC levels
 10 between 100 and 50 $\mu\text{eq/L}$, the fitness of sensitive species (e.g., brook trout, zooplankton) also
 11 begins to decline; however, the overall health of the community remains good. When ANC
 12 concentrations are <50 $\mu\text{eq/L}$, they are generally associated with death or loss of fitness of biota
 13 that are sensitive to negative effects on biota that are sensitive to acidification (Kretser et al.,
 14 1989; Dennis and Bulger, 1995).



15
 16 **Figure 4.2-1. (a)** Number of fish species per lake or stream versus acidity,
 17 expressed as acid neutralizing capacity for Adirondack Case Study Area lakes
 18 (Sullivan et al., 2006). **(b)** Number of fish species among 13 streams in
 19 Shenandoah National Park. Values of acid neutralizing capacity are means based
 20 on quarterly measurements from 1987 to 1994. The regression analysis shows a
 21 highly significant relationship ($p < .0001$) between mean stream acid neutralizing
 22 capacity and the number of fish species.

23 When ANC concentrations drop to <20 $\mu\text{eq/L}$, all biota exhibit some level of negative
 24 effects. Fish and plankton diversity and the structure of the communities continue to decline
 25 sharply to levels where acid-tolerant species begin to outnumber all other species (Matuszek and

1 Beggs, 1988; Driscoll et al., 2001). Stoddard et al. (2003) showed that to protect biota from
2 episodic acidification in the springtime, base flow ANC concentrations had to have an ANC of at
3 least 30-40 $\mu\text{eq/L}$ (Figure 4.1-1 of Appendix 4).

4 Complete loss of fish populations and extremely low diversity of planktonic communities
5 occur when ANC concentrations stay $<0 \mu\text{eq/L}$. Only acidophilic species are present, but their
6 population numbers are sharply reduced (Sullivan et al., 2006).

7 **4.2.1.3 Ecosystem Services**

8 Because acidification primarily affects the diversity and abundance of aquatic biota, it
9 also primarily affects the ecosystem services that are derived from the fish and other aquatic life
10 found in these surface waters.

11 **Provisioning Services.** Food and fresh water are generally the most important
12 provisioning services provided by inland surface waters (MEA, 2005). Whereas acidification is
13 unlikely to have serious negative effects on, for example, water supplies for municipal,
14 industrial, or agricultural uses, it can limit the productivity of surface waters as a source of food
15 (i.e., fish). In the northeastern United States, the surface waters affected by acidification are not a
16 major source of commercially raised or caught fish; however, they are a source of food for some
17 recreational and subsistence fishers and for other consumers. Although data and models are
18 available for examining the effects on recreational fishing, relatively little data are available for
19 measuring the effects on subsistence and other consumers. For example, although there is
20 evidence that certain population subgroups in the northeastern United States, such as the Hmong
21 and Chippewa ethnic groups, have particularly high rates of self-caught fish consumption
22 (Hutchison and Kraft, 1994; Peterson et al., 1994), it is not known if and how their consumption
23 patterns are affected by the reductions in available fish populations caused by surface water
24 acidification.

25 **Cultural Services.** Inland surface waters support several cultural services, such as
26 aesthetic and educational services; however, the type of service that is likely to be most widely
27 and significantly affected by aquatic acidification is recreational fishing. Recreational fishing in
28 lakes and streams is among the most popular outdoor recreational activities in the northeastern
29 United States. Data from the 2006 National Survey of Fishing, Hunting, and Wildlife Associated
30 Recreation (FHWAR) indicate that $>9\%$ of adults in this part of the country participate annually
31 in freshwater (excluding Great Lakes) fishing. The total number of freshwater fishing days

1 occurring in those states (by both residents and nonresidents) in 2006 was 140.8 million days.
2 Roughly two-thirds of these fishing days were at ponds, lakes, or reservoirs in these states, and
3 the remaining one-third were at rivers or streams. Based on studies conducted in the northeastern
4 United States, Kaval and Loomis (2003) estimated average consumer surplus values per day of
5 \$35.91 for recreational fishing (in 2007 dollars); therefore, the implied total annual value of
6 freshwater fishing in the northeastern United States was \$5.06 billion in 2006. Consumer surplus
7 value is a commonly used and accepted measure of economic benefit (see, for example, U.S.
8 EPA, 2000). It is the difference between (1) the maximum amount individuals are, on average,
9 willing and able to pay for a good, service, or activity (in this case, a day of recreational fishing)
10 and (2) the amount they actually pay (in out-of-pocket and time costs). For recreation days, it is
11 most commonly measured using recreation demand, travel cost models.

12 **Regulating Services.** In general, inland surface waters, such as lakes, rivers, and streams
13 provide a number of regulating services, such as hydrological regime regulation and climate
14 regulation. There is little evidence that acidification of freshwaters in the northeastern United
15 States has significantly degraded these specific services; however, freshwater ecosystems also
16 provide biological control services by providing environments that sustain delicate aquatic food
17 chains. These services are certainly disrupted by the toxic effects of acidification on fish and
18 other aquatic life. Although it is difficult to quantify these services and how they are affected by
19 acidification, it is worth noting that some of these services may be captured through measures of
20 provisioning and cultural services. For example, these biological control services may serve as
21 “intermediate” inputs that support the production of “final” recreational fishing and other cultural
22 services.

23 **4.2.2 Characteristics of Sensitive Areas**

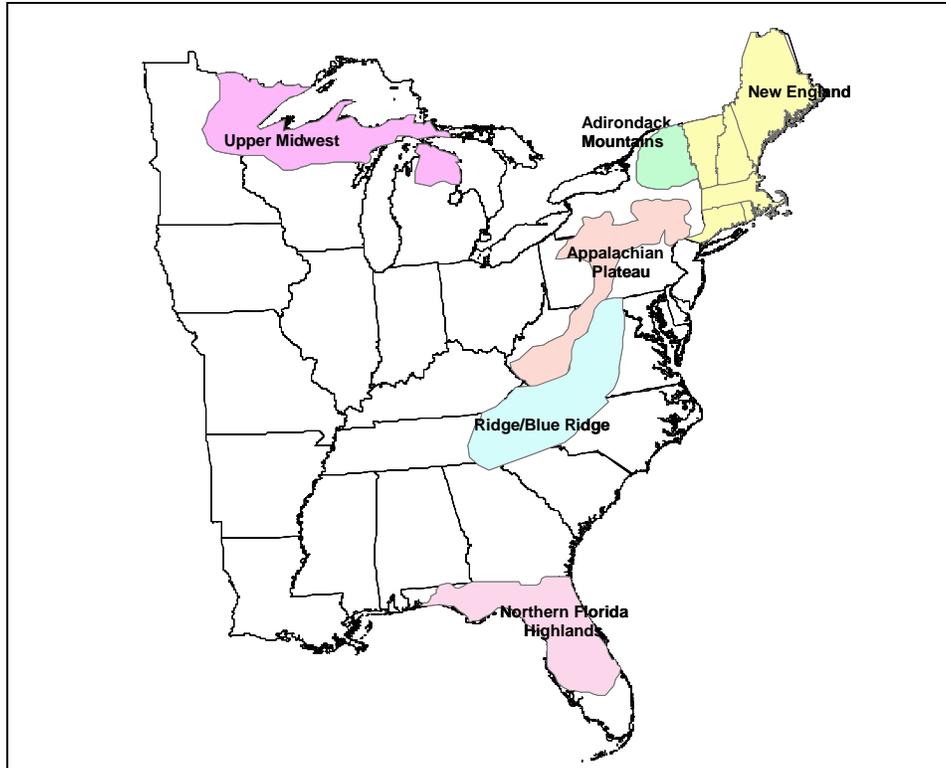
24 The ISA reports that the principal factor governing the sensitivity of terrestrial and
25 aquatic ecosystems to acidification from sulfur and nitrogen deposition is geology (particularly
26 surficial geology). Geologic formations having low base cation supply generally underlie the
27 watersheds of acid-sensitive lakes and streams. Other factors that contribute to the sensitivity of
28 soils and surface waters to acidifying deposition include topography, soil chemistry, land use,
29 and hydrologic flowpath. Surface waters in the same setting can have different sensitivities to
30 acidification, depending on the relative contributions of near-surface drainage water and deeper
31 groundwater (Chen et al., 1984; Driscoll et al., 1991; Eilers et al., 1983). Lakes and streams in

1 the United States that are sensitive to episodic and chronic acidification in response to SO_x, and
2 to a lesser extent NO_x, deposition tend to occur at relatively high elevation in areas that have
3 base-poor bedrock, high relief, and shallow soils (U.S. EPA, 2008, Section 3.2.4.1).

4 The regions of the United States with low surface water ANC values are the areas that are
5 sensitive to acidifying deposition. The majority of lakes and streams in the United States have
6 ANC levels >200 µeq/L and are not sensitive to the deposition of NO_x and SO_x air pollution at
7 their existing ambient concentration levels. **Figure 4.2-2** shows the acid-sensitive regions of the
8 eastern United States with the potential of low surface water ANC, as determined by geology and
9 surface water chemistry.

10 Freshwater surveys and monitoring in the eastern United States have been conducted by
11 many programs since the mid-1980s, including EPA's Environmental Monitoring and
12 Assessment Program (EMAP), National Surface Water Survey (NSWS), Temporally Integrated
13 Monitoring of Ecosystems (TIME) (Stoddard, 1990), and Long-term Monitoring (LTM) (Ford et
14 al., 1993; Stoddard et al., 1998) programs. Based on surface water data from these programs,
15 New England, the Adirondack Mountains, the Appalachian Mountains (northern Appalachian
16 Plateau and Ridge/Blue Ridge region), northern Florida, and the Upper Midwest contain the
17 most sensitive lakes and streams (i.e., ANC less than about 50 µeq/L) since the 1980s.

18 New England, the Adirondack Mountains, the northern Appalachian Plateau, the
19 Ridge/Blue Ridge region, and the Upper Midwest are estimated to contain 95% of the lakes and
20 84% of the streams in the United States that have been anthropogenically acidified through
21 deposition. In 2002, Stoddard et al. (2003) took another comprehensive look at the level of
22 acidification within all of these regions. Although improvement in ANC occurred, about 8% of
23 lakes in the Adirondack Mountains and 6% to 8% of streams in the northern Appalachian Plateau
24 and Ridge/Blue Ridge region were still acidic at base-flow conditions. Because they are still
25 receiving substantial NO_x/SO_x deposition inputs and still contain a large number of waterbodies
26 that are acidic, areas in New England, the Adirondack Mountains, the Northern Appalachian
27 Plateau, and the Ridge/Blue Ridge region provide ideal case study areas to assess the risk to
28 aquatic ecosystems from NO_x/SO_x acidifying deposition.



1
2 **Figure 4.2-2.** Ecosystems sensitive to acidifying deposition in the eastern United
3 States (U.S. EPA, modified from NAPAP, 2005).

4 **4.2.3 Case Study Area Selection**

5 Selection of case study areas was based on **Figure 4.2-2** (showing areas of the potential
6 sensitivity to aquatic acidification), potential case study areas identified in the ISA (U.S. EPA,
7 2008, Table 4-4), and sites recommended for consideration by the Ecological Effects
8 Subcommittee (EES) of the Advisory Council for Clean Air Compliance Analysis (U.S. EPA,
9 2005). After considering this information, the Adirondack Mountains and the Shenandoah
10 Mountains (referred to in this chapter as Adirondack and Shenandoah case study areas,
11 respectively) were selected. The rationale for choosing these two case study areas is described in
12 the following subsections.

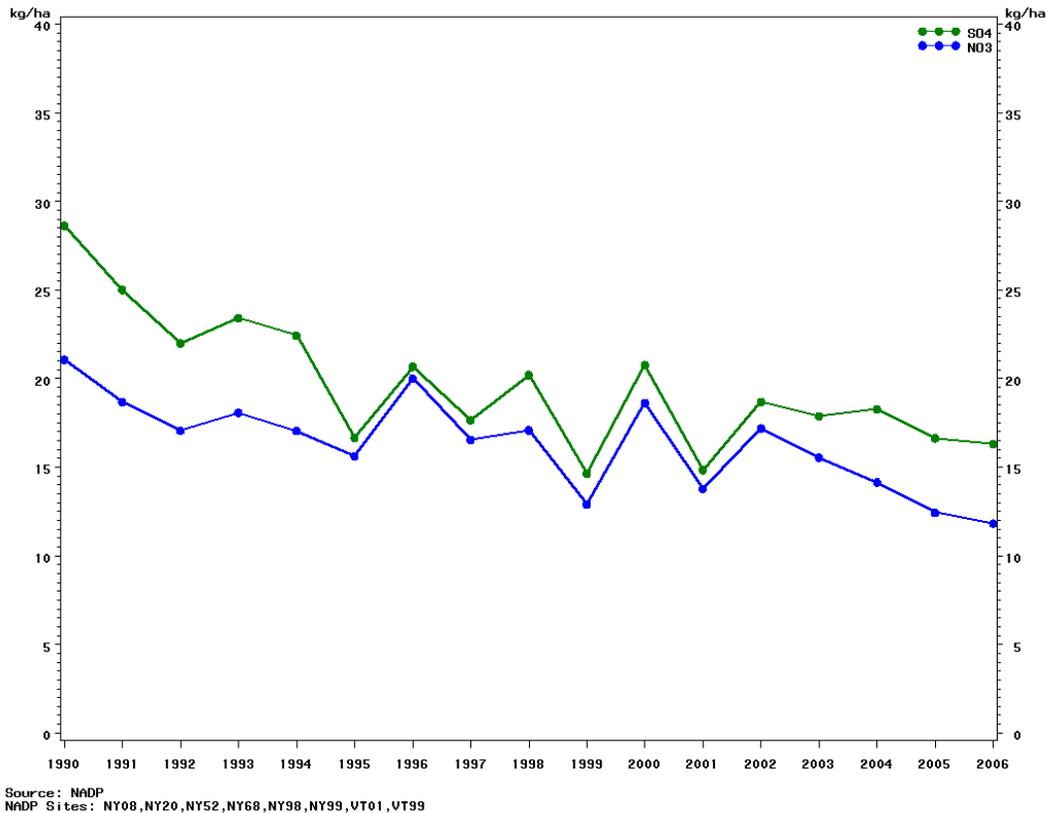
13 **4.2.3.1 Adirondack Case Study Area**

14 The Adirondack Case Study Area is situated in northeastern New York and is
15 characterized by dense forest cover and abundant surface waters, with 46 peaks that extend up to
16 1600 meters (m) in elevation. The case study area includes the headlands of five major drainage
17 basins: Lake Champlain and the Hudson, Black, St. Lawrence, and Mohawk rivers, which all

1 draw water from the preserve. There are more than 2,800 lakes and ponds, and more than 1,500
2 miles of rivers that are fed by an estimated 30,000 miles of brooks and streams.

3 The Adirondack Case Study Area, particularly its southwestern section, is sensitive to
4 acidifying deposition because it receives high precipitation amounts with high concentrations of
5 pollutants, has shallow base-poor soils, and is underlain by igneous bedrock with low weathering
6 rates and buffering ability (Driscoll et al., 1991; Sullivan et al., 2006). The Adirondack Case
7 Study Area is among the most severely acid-impacted regions in North America (Driscoll et al.,
8 2003; Landers et al., 1988; Stoddard et al., 2003). It has long been used as an indicator of the
9 response of forest and aquatic ecosystems to changes in emissions of sulfur dioxide (SO₂) and
10 NO_x resulting, in part, from the Clean Air Act Amendments of 1990 (NAPAP, 1998; U.S. EPA,
11 1995).

12 Wet deposition in the Adirondack Case Study Area has been monitored by the National
13 Atmospheric Deposition Program/National Trends Network (NADP/NTN) since 1978 at two
14 sites (i.e., Huntington Forest and Whiteface Mountain) and since the 1980s at seven other sites.
15 Since 1990, wet SO₄⁻sulfate and NO₃⁻ deposition at these NADP/NTN sites in the Adirondack
16 Case Study Area has declined by about 45% and 40%, respectively (**Figure 4.2-3**). However,
17 annual total wet deposition is still more than 15 and 10 kilograms/hectare/year (kg/ha/yr) of
18 SO₄²⁻ and NO₃⁻, respectively.



Source: NADP
NADP Sites: NY08, NY20, NY52, NY68, NY98, NY99, VT01, VT99

1
2 **Figure 4.2-3.** Annual average total wet deposition (kg/ha/yr) for the period 1990
3 to 2006 in SO₄²⁻ (green) and NO₃⁻ (blue) from eight NADP/NTN sites in the
4 Adirondack Case Study Area.

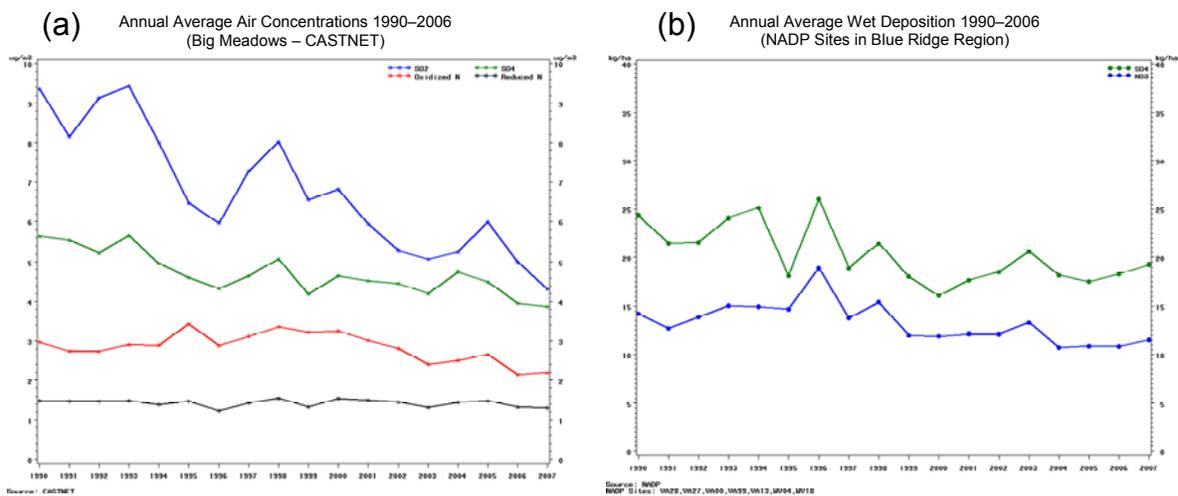
5 **4.2.3.2 Shenandoah Case Study Area**

7 The Shenandoah Case Study Area straddles
9 the crest of the Blue Ridge Mountains in western
11 Virginia, on the eastern edge of the central
13 Appalachian Mountain region. Several areas in
15 Shenandoah National Park have been designated
17 Class 1 Wilderness areas. Shenandoah National
19 Park is known for its scenic beauty, outstanding
21 natural features, and biota. Air pollution within the
23 Shenandoah Case Study Area, including
25 concentrations of sulfur, nitrogen, and ozone (O₃),
26 is higher than in most other national parks in the United States.

“Bulger et al. (2000) predicted that future losses of native brook trout (*Salvelinus fontinalis*) populations in the streams of western Virginia will be substantial unless acidic deposition reductions are much greater than the 1990 Clean Air Act Amendments will provide...Despite recent declines in acidic deposition and some encouraging evidence for initial recovery in other parts of the country, recovery in the central Appalachian region in general, and the Shenandoah National Park in particular, has been limited and impairment of surface waters due to acidic deposition continues (Stoddard et al. 2003; Webb et al. 2004).” (Webb, 2004)

1 This area is sensitive to acidifying deposition because it receives high precipitation, has
 2 shallow base-poor soils, and is underlain by igneous and silicon (Si)-based bedrock with low
 3 weathering rates and poor ANC. The Shenandoah Case Study Area is also among the most
 4 severely acid-impacted regions in North America (Stoddard et al., 2003; Webb et al., 2004).

5 Wet deposition in the Shenandoah National Park monitored at 7 sites by the NADP/NTN
 6 since the 1980s shows wet SO_4^{2-} and NO_3^- deposition declining by about 28% and 20%,
 7 respectively (**Figure 4.2-4 a, b**). However, annual total deposition is still over 15 and 10 kg/ha/yr
 8 of SO_4^{2-} and NO_3^- , respectively.



9
 10 **Figure 4.2-4.** Air pollution concentrations and deposition for the period 1990 to
 11 2006 using one CASTNET and seven NADP/NTN sites in the Shenandoah Case
 12 Study Area. **(a)** Annual average air concentrations of SO_2 (blue), oxidized
 13 nitrogen (red), SO_4^{2-} (green), and reduced nitrogen (black). **(b)** Annual average
 14 total wet deposition (kg/ha/yr) of SO_4^{2-} (green) and NO_3^- (blue).

15 4.2.4 Current Conditions in Case Study Areas

16 4.2.4.1 Approach

17 Status of current conditions and trends in SO_4^{2-} , NO_3^- , and ANC concentrations measured
 18 in surface water were used to characterize links to the effects of acidifying deposition on the
 19 acid-base chemistry of a waterbody. Trends in these sensitive chemical receptors show whether
 20 the conditions of a waterbody are improving and heading toward recovery or are continuing to
 21 degrade.

1 **MAGIC Modeling and Input Data.** To assess surface water trends in SO_4^{2-} , NO_3^- , and
2 ANC concentrations, surface water monitoring data from the EPA-administered LTM program
3 was used (see Appendix 4's Attachment 4.B for more details on TIME/LTM network). Trends in
4 SO_4^{2-} , NO_3^- , and ANC concentrations were assessed using average yearly values for the period
5 from 1990 to 2006.

6 The preacidification condition of a waterbody is rarely known because it can no longer be
7 measured. Likewise, it is also difficult to determine whether a waterbody has recovered or will
8 recover from acidification as acidifying deposition inputs decline, because recovery may take
9 many years to occur. For these reasons, hydrological models, such as MAGIC, enable estimates
10 of past, present, and future water quality levels that can be used to evaluate (1) the associated
11 risk and uncertainty of the current levels of acidification as compared with preacidification
12 conditions, and (2) whether a system will recover as a result of a reduction in acidifying
13 deposition.

14 MAGIC was used to determine the past (preacidification), present (2002 and 2008), and
15 future (2020 and 2050) acidic conditions of 44 lakes in the Adirondack Case Study Area and 60
16 streams in the Shenandoah Case Study Area (**Figure 4.2-5**). Furthermore, MAGIC was used to
17 evaluate the associated risk and uncertainty of the current levels of acidification given the pre-
18 acidification water quality and the levels of uncertainty in the input parameters. The MAGIC
19 model output for each waterbody was summarized into five ANC levels that correspond to the
20 aquatic status categories *Acute Concern*, *Severe Concern*, *Elevated Concern*, *Moderate Concern*,
21 and *Low Concern*. This grouping offers an assessment of the current risk to the biota of current
22 condition compared to preacidification and future conditions. Surface water chemistry data were
23 used from two EPA-administered surface water monitoring and survey programs: the TIME and
24 the LTM programs. Average yearly ANC concentrations were calculated from annual
25 measurements.



1 **Figure 4.2-5.** (Top) The location of lakes in the Adirondack Case Study Area
 2 used for **MAGIC** (red dots) and critical load (green dots) modeling sites.
 3 (Bottom) The location of streams used for both **MAGIC** and critical load
 4 modeling for the Shenandoah Case Study Area.

5 **Connecting current total nitrogen and sulfur deposition to acid-base conditions of**
 6 **lakes and streams: The Critical Load approach.** The critical load approach was used to
 7 connect current deposition of nitrogen and sulfur to the acid-base condition and biological risk to
 8 biota of lakes and streams in the study. Calculating critical load exceedances (i.e., the amount of
 9 deposition above the critical load) allows the determination of whether current deposition poses a
 10 risk of acidification to a given group of waterbodies. This approach also allows for the
 11 comparison of different levels of ANC thresholds (e.g., 0, 20, 50, 100 $\mu\text{eq/L}$) and their associated

1 risk to the biological community. **Table 4.2-1** provides a summary of the biological effects
2 experienced at each of these limits.

4 Critical loads and their exceedances at four levels
6 of biological protection were calculated for 169 lakes in
8 the Adirondack Case Study Area and 60 streams in the
10 Shenandoah Case Study Area. Four ANC limits (i.e.,
12 ANC_{limit}) of biological protection were used: 0 µeq/L
14 (low protection), 20 µeq/L (minimal protection), 50
16 µeq/L (moderate protection), and 100 µeq/L (full
18 protection). A full and complete description of the
20 biological effects at a given ANC limit appears in
22 Appendix 4, Section 4.1.

24 From the 169 modeled lakes and 60 streams in
26 the Adirondack and Shenandoah case study areas,
28 respectively, the number and percentage of waterbodies
30 that receive acidifying deposition above their critical
31 loads for a given ANC limit of 0, 20, 50, and 100 µeq/L were determined.

The **critical load approach** provides a means of gauging whether a group of lakes or streams in a given area receives deposition that results in a level of biological harm that is defined by an ANC concentration, known as the critical limit, which corresponds to harmful biological effects (e.g., ANC of 50 µeq/L). A critical load estimate is analogous to determining the “susceptibility” of a waterbody to become acidified from the deposition of nitrogen and sulfur. Low critical load values (i.e., less than 50 meq/m²·yr) mean that the watershed has a limited ability to neutralize the addition of acidic anions, and hence, it is susceptible to acidification. The greater the critical load value, the greater the ability of the watershed to neutralize the additional acidic anions and protect aquatic life.

Table 4.2-1. Aquatic Status Categories

Category Label	ANC Levels*	Expected Ecological Effects
Acute Concern	<0 micro equivalent per Liter (µeq/L)	Complete loss of fish populations is expected. Planktonic communities have extremely low diversity and are dominated by acidophilic forms. The number of individuals in plankton species that are present is 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. 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, reproduction capacity, and fitness. Diversity and distribution of zooplankton communities decline.

Category Label ANC Levels* Expected Ecological Effects		
Moderate Concern	50–100 $\mu\text{eq/L}$	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 $\mu\text{eq/L}$	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.

1

2 4.2.4.2 Current Conditions in Adirondack Case Study Area Surface Waters

3 **Current and preacidification conditions of surface waters.** Since the mid-1990s, lakes

4 in the Adirondack Case Study Area have
5 shown signs of improvement in NO_3^- and
6 SO_4^{2-} concentrations in surface waters. Wet
7 deposition rates for SO_2 and NO_x have been
8 reduced (**Figure 4.2-3**), and, as a result, NO_3^-
9 and SO_4^{2-} concentrations have decreased in
10 surface waters by approximately 26% and
11 13%, respectively (**Figure 4.2-6**).

12 The decline in SO_4^{2-} concentrations in
13 surface waters in the Adirondack Case Study

14 Area is $-2.1 \mu\text{eq/L/year}$, while the decline in NO_3^- is $-0.23 \mu\text{eq/L/year}$. However, current
15 concentrations of NO_3^- and SO_4^{2-} are still well above preacidification conditions based on

17 MAGIC model simulations. **Figure 4.2-7** and **Figure 4.2-8** show
19 the condition of the lakes in 1860 “preacidification” and in 2006
21 “current” conditions. On average, NO_3^- and SO_4^{2-} concentrations
23 are 17- and 5-fold higher today, respectively (**Table 4.2-2**).

Table 4.2-2. Estimated Average Concentrations of Surface Water Chemistry at 44 Lakes in the Adirondack Case Study Area Modeled Using MAGIC for Preacidification (1860) and Current (2006) Conditions

ueq/L	Preacidification		Current	
	Avg.	(+/-)	Avg.	(+/-)
ANC	120.3	13.6	62.1	15.7
SO_4^{2-}	12.4	2.1	66.1	1.24
NO_3^-	0.2	1.7	3.4	14.8
NH_4^+	0.0	0.0	0.1	0.1

Current NO_3^- and SO_4^{2-} concentrations are 17- and 5-fold higher in Adirondack Case Study Area lakes today than they were in 1860.

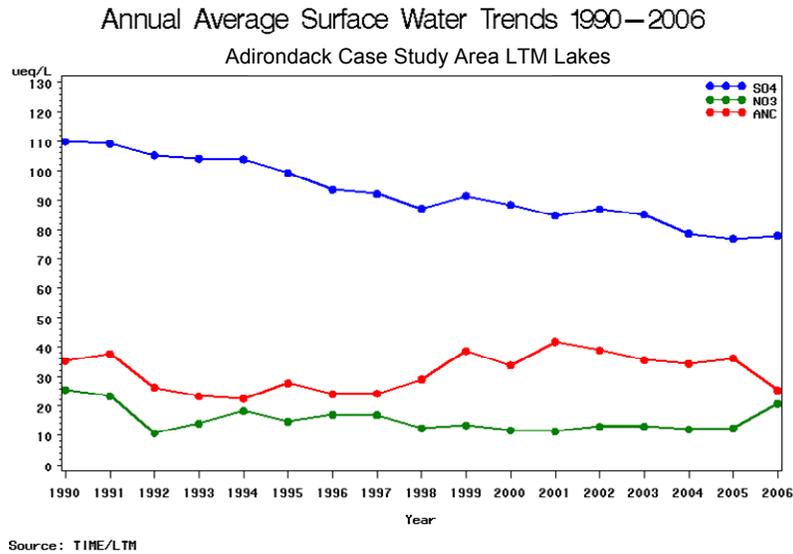


Figure 4.2-6. Trends over time for SO_4^{2-} , NO_3^- , and acid neutralizing capacity in LTM. SO_4^{2-} and NO_3^- concentrations have decreased in surface waters by approximately 26% and 13%, respectively.

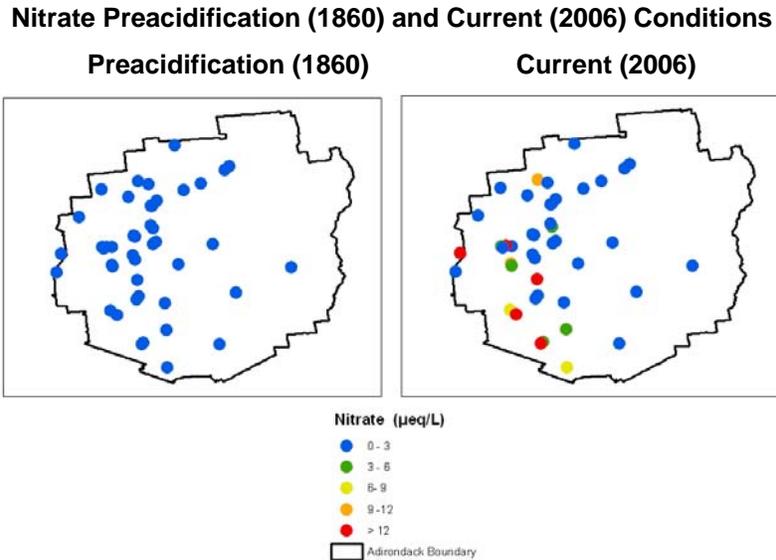
Although NO_3^- deposition can be an important factor in acid precipitation, these current results demonstrate that acidification in the Adirondack Case Study Area is currently being driven by SO_4^{2-} deposition because the current average SO_4^{2-} concentration is some 19-fold greater than NO_3^- concentrations in surface waters (**Table 4.2-2**).

An increase in ANC concentrations of +1 µeq/L/year has corresponded to the declines in NO_3^- and SO_4^{2-} , despite reductions in base cations of Ca^{2+} and Mg^{2+} during the same period of time. This decline in base cation concentration is important because base cations buffer the inputs of NO_3^- and SO_4^{2-} , which will likely limit future recovery of ANC concentrations. In the Adirondack Case Study Area, levels of Al also declined slightly (data not shown).

Based on the observed annual average concentration of ANC, there is still a significant number of lakes in the Adirondack Case Study Area that have *Elevated* (i.e., ANC <50 µeq/L) to *Severe* (i.e., ANC <20 µeq/L) condition of acidity (**Figure 4.2-9**).

Based on monitoring data, only 22% of monitored lakes are “not acidic,” which include the *Moderate to Low Concern* classes, and thus have water quality that poses little risk to aquatic biota. On the other hand, 78% of all monitored lakes have a current risk of *Elevated*, *Severe*, or *Acute*. Of that 78%, 31% experience episodic acidification (i.e., severe concern) and 18% are chronically acidic today (i.e., acute concern).

1
2

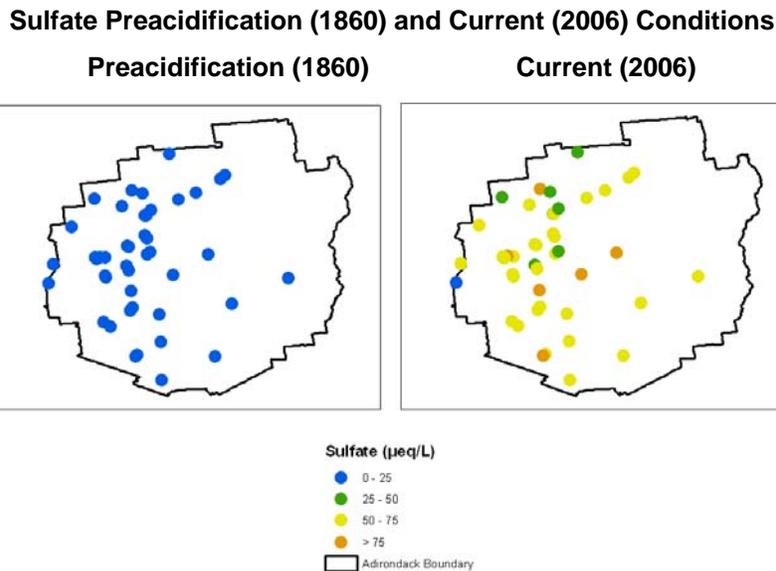


3
4
5
6

Source: EPA/CAMD 2009

Figure 4.2-7. NO_3^- concentrations of preacidification (1860) and current (2006) conditions based on hindcasts of 44 lakes in the Adirondack Case Study Area modeled using MAGIC.

7
8



9
10
11
12

Source: EPA/CAMD 2009

Figure 4.2-8. SO_4^{2-} concentrations of preacidification (1860) and current (2006) conditions based on hindcasts of 44 lakes in the Adirondack Case Study Area modeled using MAGIC.

1 An estimate of the level of current condition
 2 at these lakes that can be attributed to the effects of
 3 industrially generated acidifying deposition can be
 4 made by examining the hindcast conditions of the
 5 lakes derived from the MAGIC model output. Based
 6 on these simulations, preacidification average ANC
 7 concentration of 44 modeled lakes is 120.3 ± 13.6
 8 $\mu\text{eq/L}$, as compared with $62.1 \pm 15.7 \mu\text{eq/L}$ for today
 9 (Table 4.2-2). Furthermore, 89% of the modeled
 10 lakes are likely “not acidic” prior to the onset of
 11 acidifying deposition (Figure 4.2-10 and Figure 4.2-
 12 11). The other 11% of lakes have ANC of $>20 \mu\text{eq/L}$.
 13 The hindcast simulations produced no lakes with
 14 *Acute* or *Severe Concern* preacidification condition,
 15 suggesting that current ambient concentrations of
 16 NO_x and SO_x and their associated levels of NO_3^- and
 17 SO_4^{2-} deposition pose a risk of acidification to
 18 approximately 32% of modeled lakes.

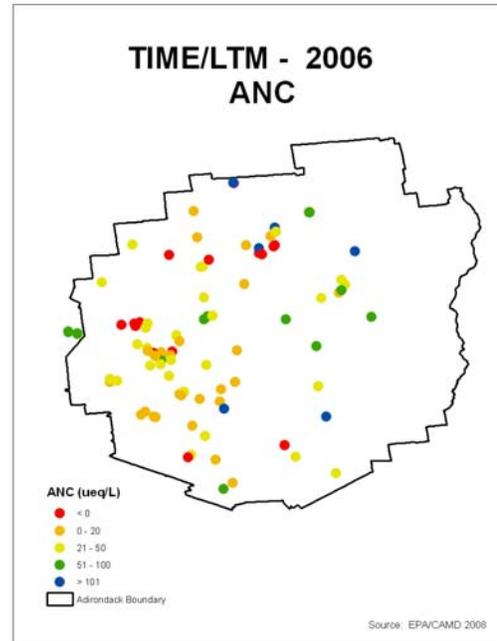
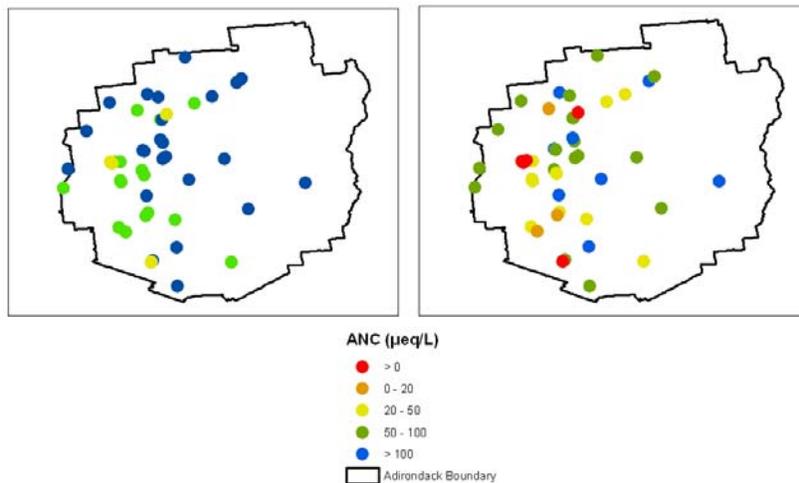
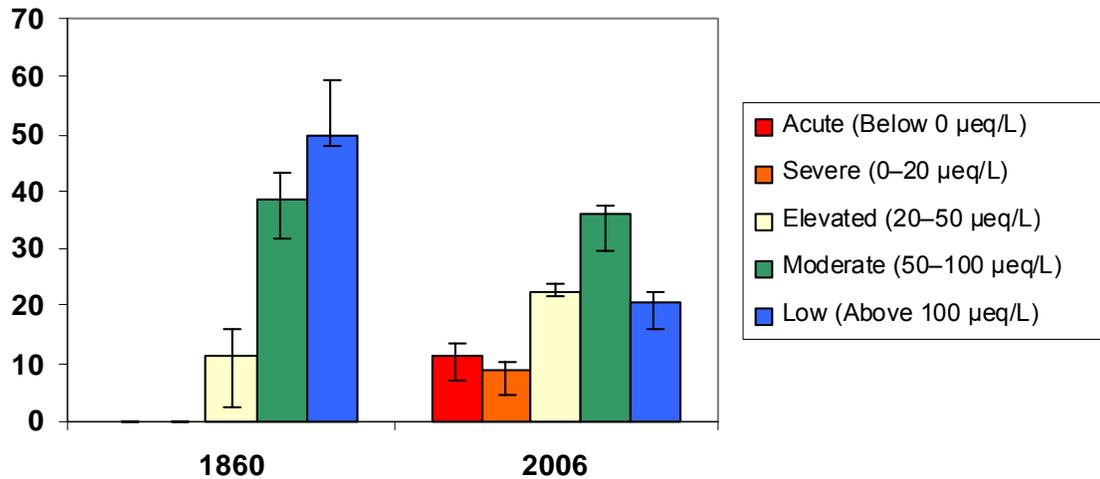


Figure 4.2-9. Acid neutralizing capacity concentrations from 94 lakes in the Adirondack Case Study Area. Monitoring data from the TIME/LTM programs.

19 **ANC Preacidification (1860) and Current (2006) Conditions**
 20 **Preacidification (1860) Current (2006)**



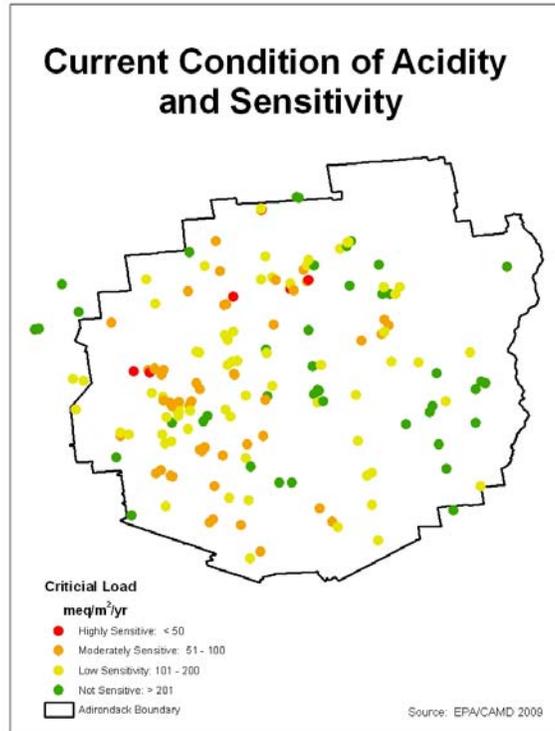
21 Source: EPA/CAMD 2009
 22 **Figure 4.2-10.** Acid neutralizing capacity concentrations of preacidification
 23 (1860) and current (2006) conditions based on hindcasts of 44 modeled lakes in
 24 the Adirondack Case Study Area.



1
2 **Figure 4.2-11.** Percentage of Adirondack Case Study Area lakes in the five
3 classes of acidification (i.e., Acute, Severe, Elevated, Moderate, Low) for years
4 2006 and 1860 (preacidification) for 44 lakes modeled using MAGIC. Error bar
5 indicates the 95% confidence interval.

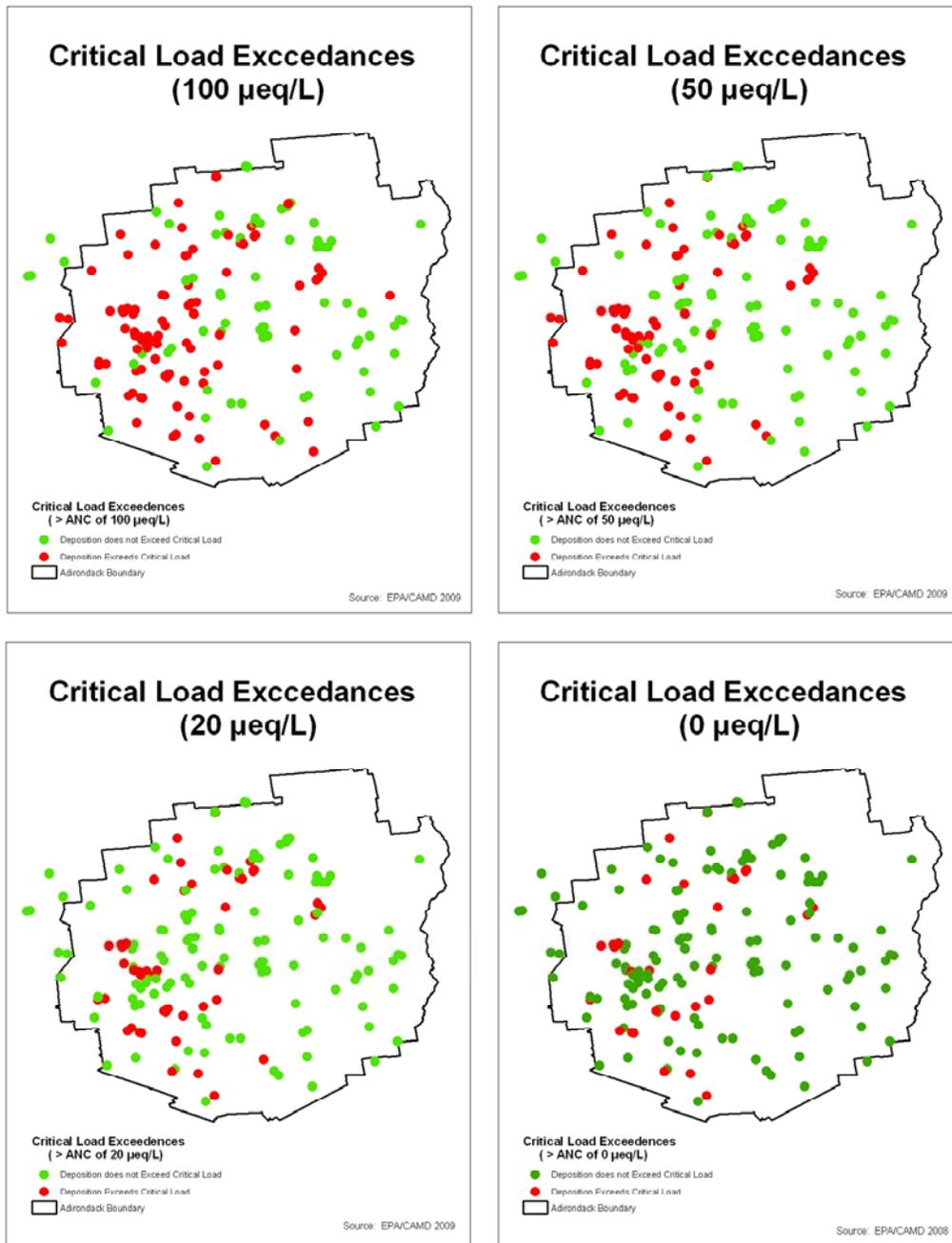
6 **The biological risk from current total nitrogen and sulfur deposition: Critical load**
7 **assessment.** In **Figure 4.2-12**, a critical load indicates the amount of acidic input of total sulfur
8 and nitrogen deposition that a lake can neutralize and still maintain an ANC of 50 µeq/L. Sites
9 labeled by red or orange dots have less buffering ability than sites labeled with yellow and green
10 dots, and hence, indicate those lakes that are most sensitive to acidifying deposition, due to a
11 host of environmental factors. Approximately 50% of the 169 lakes modeled in the Adirondack
12 Case Study Area are sensitive or at risk to acidifying deposition.

13 In **Figure 4.2-13**, a critical load exceedance “value” indicates combined total sulfur and
14 nitrogen deposition in year 2002 that is greater than the amount of deposition the lake could
15 buffer and still maintain the ANC level above each of the four different ANC limits of 0, 20, 50,
16 and 100 µeq/L. For the year 2002, 18%, 28%, 44%, and 58% of the 169 lakes modeled received
17 levels of combined total sulfur and nitrogen deposition that exceeded their critical load with
18 critical limits of 0, 20, 50, and 100 µeq/L, respectively (**Table 4.2-3**).



1
2
3
4
5
6
7

Figure 4.2-12. Critical loads of acidifying deposition that each surface waterbody in the Adirondack Case Study Area can receive while maintaining or exceeding an acid neutralizing capacity concentration of 50 $\mu\text{eq/L}$ based on 2002 data. Watersheds with critical load values $<100 \text{ meq/m}^2/\text{yr}$ (red and orange dots) are most sensitive to surface water acidification, whereas watersheds with values $>100 \text{ meq/m}^2/\text{yr}$ (yellow and green dots) are the least sensitive sites.



1
2 **Figure 4.2-13.** Critical load exceedances (red dots) based on 2002 deposition
3 magnitudes for Adirondack Case Study Area waterbodies where the critical limit
4 acid neutralizing capacity is 0, 20, 50, and 100 µeq/L, respectively. Green dots
5 represent lakes where current total nitrogen and sulfur deposition is below the
6 critical load. See **Table 4.2-3**.

7 **Recovery from acidification given current emission reductions.** In considering the
8 future responses of lakes to current emissions and given the current condition of the lakes, the
9 question becomes whether lakes can recover to healthy systems (i.e., ANC > 50 µeq/L), or

1 whether additional reductions in acidifying deposition is needed? The forecast model runs using
 2 MAGIC were used to determine whether current deposition could lead to recovery of the
 3 acidified lakes.

4 Based on a
 5 deposition scenario that
 6 maintains current
 7 emission levels to 2020
 8 and 2050, the simulation
 9 forecast indicates no
 10 improvement in water
 11 quality. The percentage
 12 of lakes within the

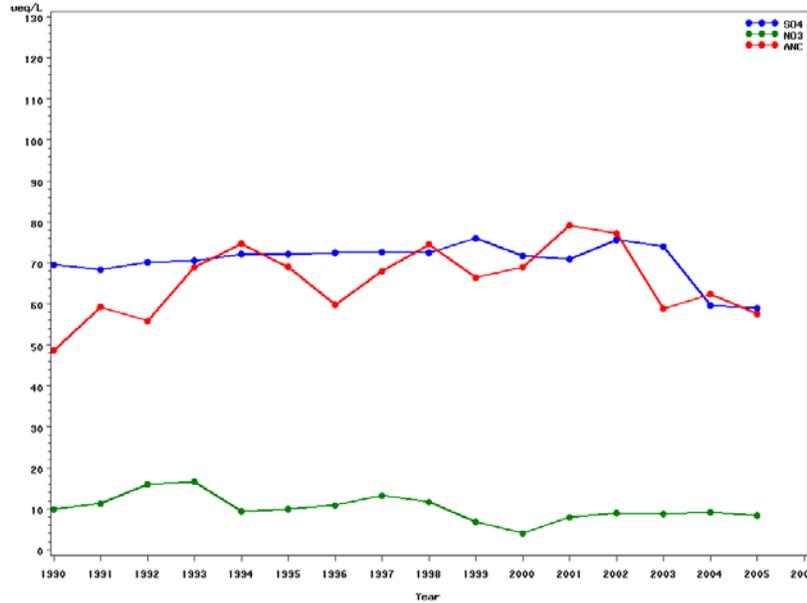
Table 4.2-3. Critical Load Exceedances (Nitrogen + Sulfur Deposition > Critical Load) for 169 Modeled Lakes Within the TIME/LTM and EMAP Survey Programs. “No. Lakes” Indicates the Number of Lakes at the Given Acid Neutralizing Capacity Limit; “% Lakes” Indicates the Total Percentage of Lakes at the Given Acid Neutralizing Capacity Limit

ANC Limit 100 µeq/L		ANC Limit 50 µeq/L		ANC Limit 20 µeq/L		ANC Limit 0 µeq/L	
No. Lakes	% Lakes	No. Lakes	% Lakes	No. Lakes	% Lakes	No. Lakes	% Lakes
98	58	74	44	47	28	30	18
Lake No. = 169							

13 *Elevated to Acute Concern* classes remains the same in 2020 and 2050. Moreover, the percentage
 14 of modeled lakes classified as “not acidic” remains the same, suggesting that current emission
 15 levels will likely not improve the acidification of lakes in the Adirondack Case Study Area. It is
 16 possible to conduct additional modeling and estimate the acidification impacts of lower
 17 emissions and alternate deposition scenarios; however, this has not been done at this time.

18 **4.2.4.3 Current Conditions in Shenandoah Case Study Area Surface Waters**

19 **Current and preacidification conditions of surface waters.** Since the mid-1990s,
 20 streams in the Shenandoah Case Study Area have shown slight signs of improvement in NO_3^-
 21 and SO_4^{2-} concentrations in surface waters. Deposition of SO_x and NO_x has decreased, but has
 22 not resulted in much improvement in NO_3^- and SO_4^{2-} stream concentrations (**Figure 4.2-14**).
 23 However, ANC concentrations increased from the about 50 µeq/L in the early 1990 to >75 µeq/L
 24 until 2002, when ANC levels declined back to 1991 to 1992 levels (**Figure 4.2-14**). At this time,
 25 it is unclear why ANC initially improved and is now declining.



Source: TIME/LTH

Figure 4.2-14. Trends over time for SO₄²⁻ (blue), NO₃⁻ (green) and acid neutralizing capacity (red) concentrations in VTSSS LTM-monitored streams in the Shenandoah Case Study Area.

The slight decline in SO₄²⁻ concentrations in surface waters of the Shenandoah Case Study Area is -0.09 µeq/L/year, while the decline in NO₃⁻ is -0.1 µeq/L/year. Current concentrations of NO₃⁻ and SO₄²⁻ are still well above preacidification conditions based on MAGIC model simulations.

Figure 4.2-15 and **Figure 4.2-16** show the condition of the streams in 1860 (preacidification) and in 2006 (current) conditions. On average, NO₃⁻ and SO₄²⁻ concentrations are 10- and 32-fold higher today, respectively (**Table 4.2-4**).

Although NO₃⁻ deposition can be an important factor in acid precipitation, these results demonstrated that acidification in the Shenandoah Case Study Area is currently being driven by SO₄²⁻ deposition since current average SO₄²⁻ concentration is 11-fold greater than NO₃⁻ concentrations in surface waters (**Table 4.2-4**).

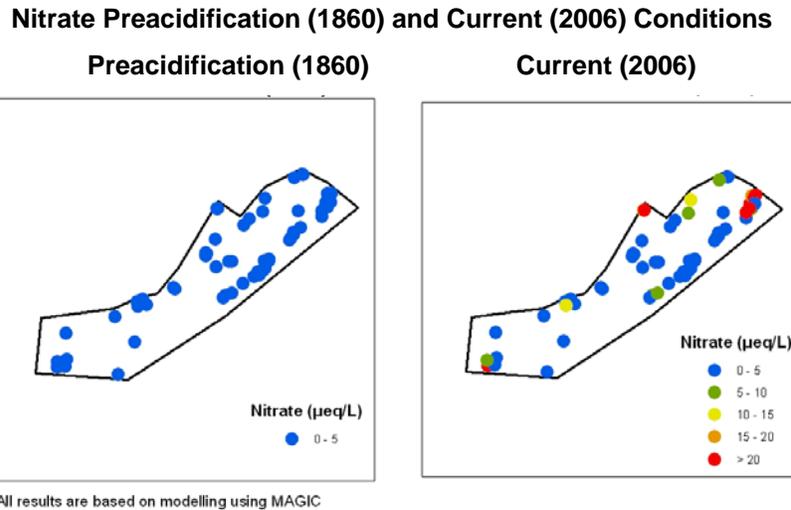
Table 4.2-4. Model Simulated Average Concentrations for Stream Chemistry at 60 Modeled Streams in the Shenandoah Case Study Area for Preacidification and Current Conditions

µeq/L	Pre-Acidification		Current	
	Avg.	(+/-)	Avg.	(+/-)
ANC	101.4	9.5	57.9	4.5
SO ₄ ²⁻	2.1	0.1	68.0	8.4
NO ₃ ⁻	0.6	0.01	6.2	0.1
NH ₄ ⁺	N/A	N/A	N/A	N/A

N/A = Not available.

Current NO₃⁻ and SO₄²⁻ concentrations are 10- and 32-fold higher in Shenandoah Case Study Area streams today than in 1860.

1 An increase in ANC concentrations of +0.08 $\mu\text{eq/L/year}$ has occurred since 1990, but for
 2 the majority of the 68 monitoring sites of the Shenandoah Case Study Area, ANC concentrations
 3 have not changed statistically from 1990 to 2006.

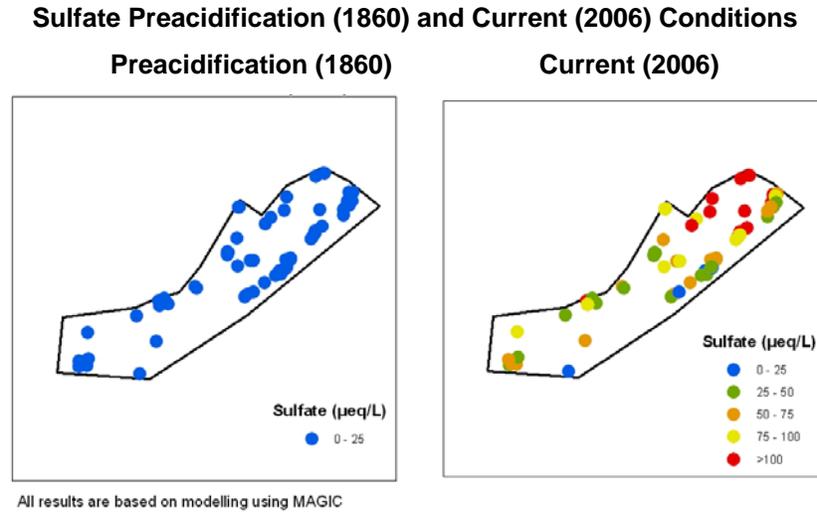


Source: EPA/CAMD 2009

7 **Figure 4.2-15.** NO_3^- concentrations of preacidification (1860) and current (2006)
 8 conditions based on hindcasts of 60 streams modeled using MAGIC in the
 9 Shenandoah Case Study Area.

10 Based on the monitored annual average for ANC, there are a significant number of
 11 streams in the Shenandoah Case Study Area that currently have *Elevated* ($\text{ANC} < 50 \mu\text{eq/L}$) to
 12 *Severe* ($\text{ANC} < 20 \mu\text{eq/L}$) classes of acidity (**Figure 4.2-17**). Only 45% of monitored streams are
 13 considered “not acidic” (i.e., of *Moderate to Low Concern*) and thus have water quality that
 14 poses less risk to aquatic biota. Approximately 55% of all monitored streams have a current risk
 15 of *Elevated, Severe, or Acute Concern*. Of that 55%, 18% experience episodic acidification
 16 (*Severe Concern*) and 12% are chronically acidic (i.e., *Acute Concern*) at current level of
 17 acidifying deposition and ambient concentration of NO_x and SO_2 .

18 An estimate of how much of this current condition is attributed to the effects of
 19 industrially generated acidifying deposition can be made by examining the hindcast conditions of
 20 the streams. Based on the MAGIC model simulations, preacidification average ANC
 21 concentration of the 60 modeled streams is $101.4 \pm 9.5 \mu\text{eq/L}$, as compared with $57.9 \pm 4.5 \mu\text{eq/L}$
 22 for today (**Table 4.2-4**).

1
2

Source: EPA/CAMD 2009

3
4
5
6

Figure 4.2-16. SO_4^{2-} concentrations of preacidification (1860) and current (2006) conditions based on hindcasts of 60 streams modeled using MAGIC in the Shenandoah Case Study Area.

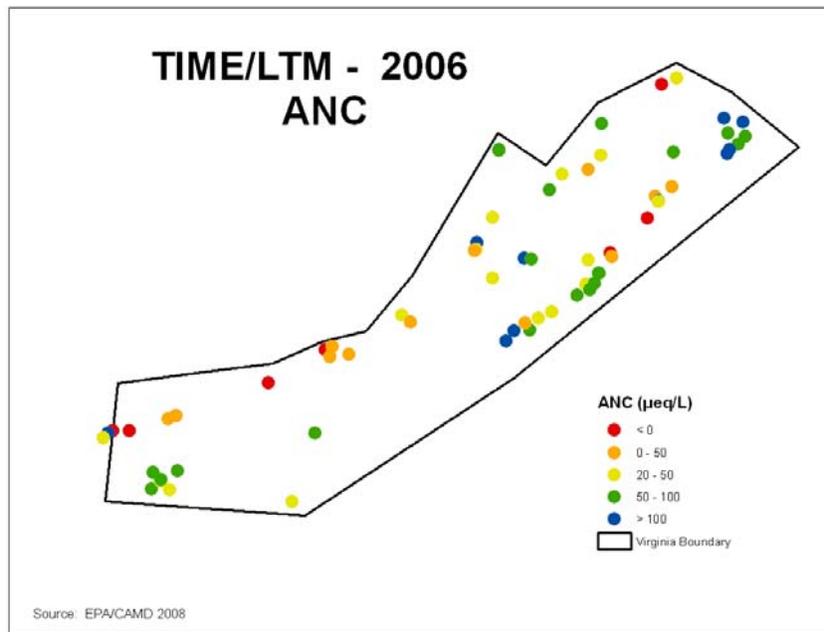
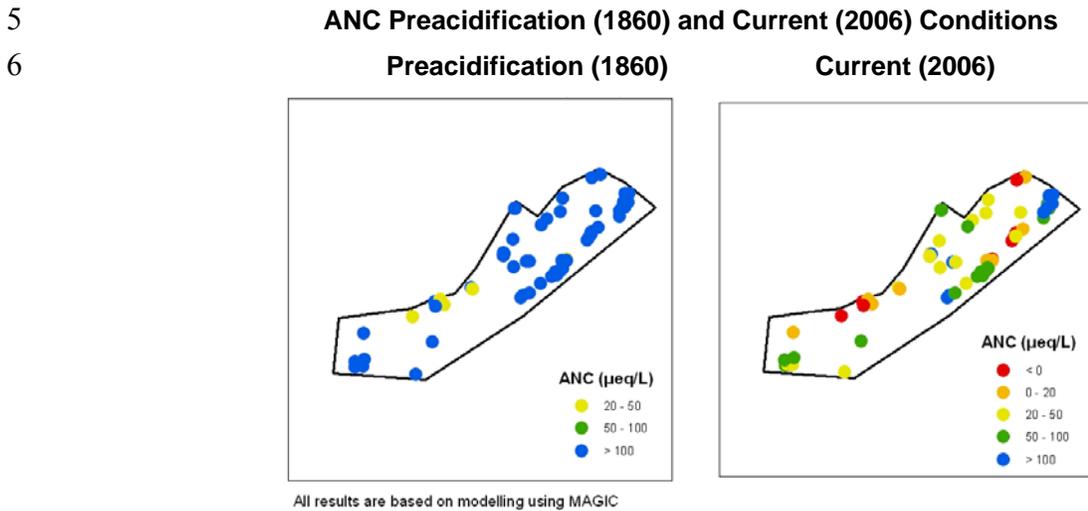
7
8
9
10

Figure 4.2-17. Acid neutralizing capacity concentrations from 68 streams in the VTSSS-SWAS/LTM monitoring network in the Shenandoah Case Study Area (2006 data).

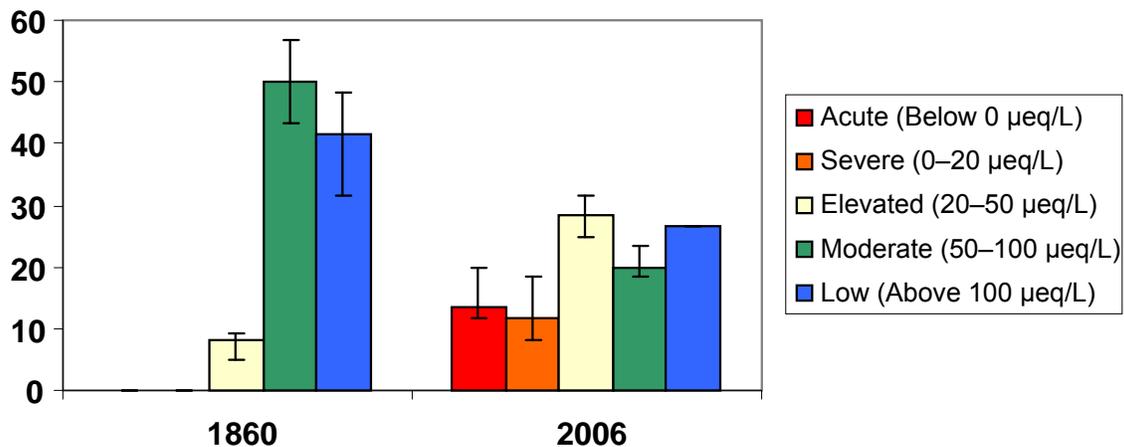
11 Furthermore, 92% of the modeled streams likely were “not acidic” prior to the onset of
12 acidifying deposition (**Figure 4.2-18** and **Figure 4.2-19**). The other 8% of streams had ANC of
13 >27 µeq/L. The hindcast simulations produced no streams with *Acute* or *Severe Concern*. These

1 results based on model reconstructions suggest that current and recent ambient concentrations of
 2 NO_3^- and SO_4^{2-} and their associated anthropogenic acidifying deposition are likely responsible
 3 for acidifying (ANC below 50 $\mu\text{eq/L}$) approximately 45% of streams modeled in the Shenandoah
 4 Case Study Area.



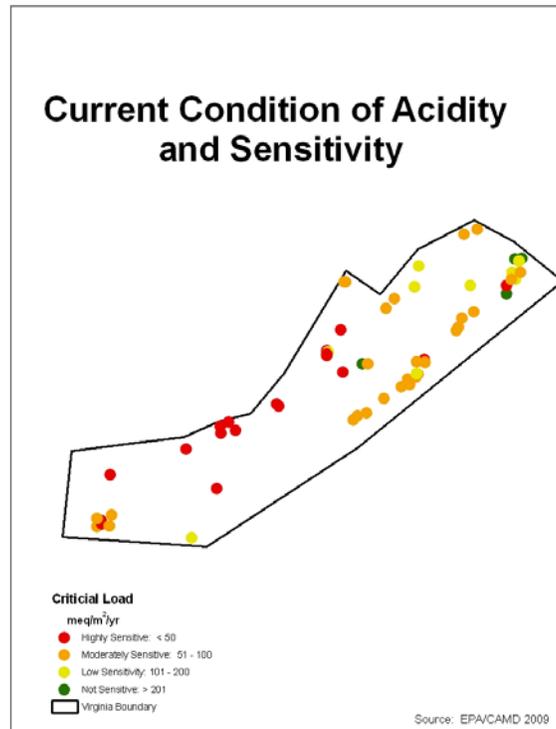
Source: EPA/CAMD 2009

7
 8 **Figure 4.2-18.** Acid neutralizing capacity concentrations of preacidification
 9 (1860) and current (2006) conditions based on hindcasts of 60 streams modeled
 10 using MAGIC in the Shenandoah Case Study Area.



11
 12 **Figure 4.2-19.** Percentage of streams in the five classes of acidification (i.e.,
 13 Acute, Severe, Elevated, Moderate, Low Concern) for years 2006 and 1860 (pre-
 14 acidification) for 60 streams modeled using MAGIC in the Shenandoah Case
 15 Study Area. The number of streams in each class is above the bar. Error bars
 16 indicate the 95% confidence interval.

1 **The biological risk from current total nitrogen and sulfur deposition: Critical load**
 2 **assessment.** In **Figure 4.2-20**, sites labeled by red or orange dots have less buffering ability than
 3 sites labeled with yellow and green dots, and hence, indicate those streams that are most
 4 sensitive to acidifying deposition, due to a host of environmental factors. Approximately 75% of
 5 the 60 streams modeled in the Shenandoah Case Study Area are sensitive or at risk to acidifying
 6 deposition.



7
 8 **Figure 4.2-20.** Critical loads of surface water acidity for an acid neutralizing
 9 capacity concentration of 50 $\mu\text{eq/L}$ for streams in the Shenandoah Case Study
 10 Area. Each dot represents an estimated amount of acidifying deposition (i.e.,
 11 critical load) that each stream's watershed can receive and still maintain a surface
 12 water acid neutralizing capacity concentration $>50 \mu\text{eq/L}$. Watersheds with
 13 critical load values $<100 \text{ meq/m}^2/\text{yr}$ (red and orange dots) are most sensitive to
 14 surface water acidification, whereas watersheds with values $>100 \text{ meq/m}^2/\text{yr}$
 15 (yellow and green dots) are the least sensitive sites.

16 In **Figure 4.2-21**, a critical load exceedance “value” indicates combined total sulfur and
 17 nitrogen deposition in year 2002 that is greater than the amount of deposition the stream could
 18 buffer and still maintain the ANC level of above each of the four different ANC limits of 0, 20,
 19 50, and 100 $\mu\text{eq/L}$. For the year of 2002, 52%, 72%, 85%, and 92% of the 60 streams modeled

- 1 receive levels of combined total sulfur and nitrogen deposition that exceeded their critical load
 2 with critical limits of 0, 20, 50, and 100 $\mu\text{eq/L}$, respectively (Table 4.2-5).

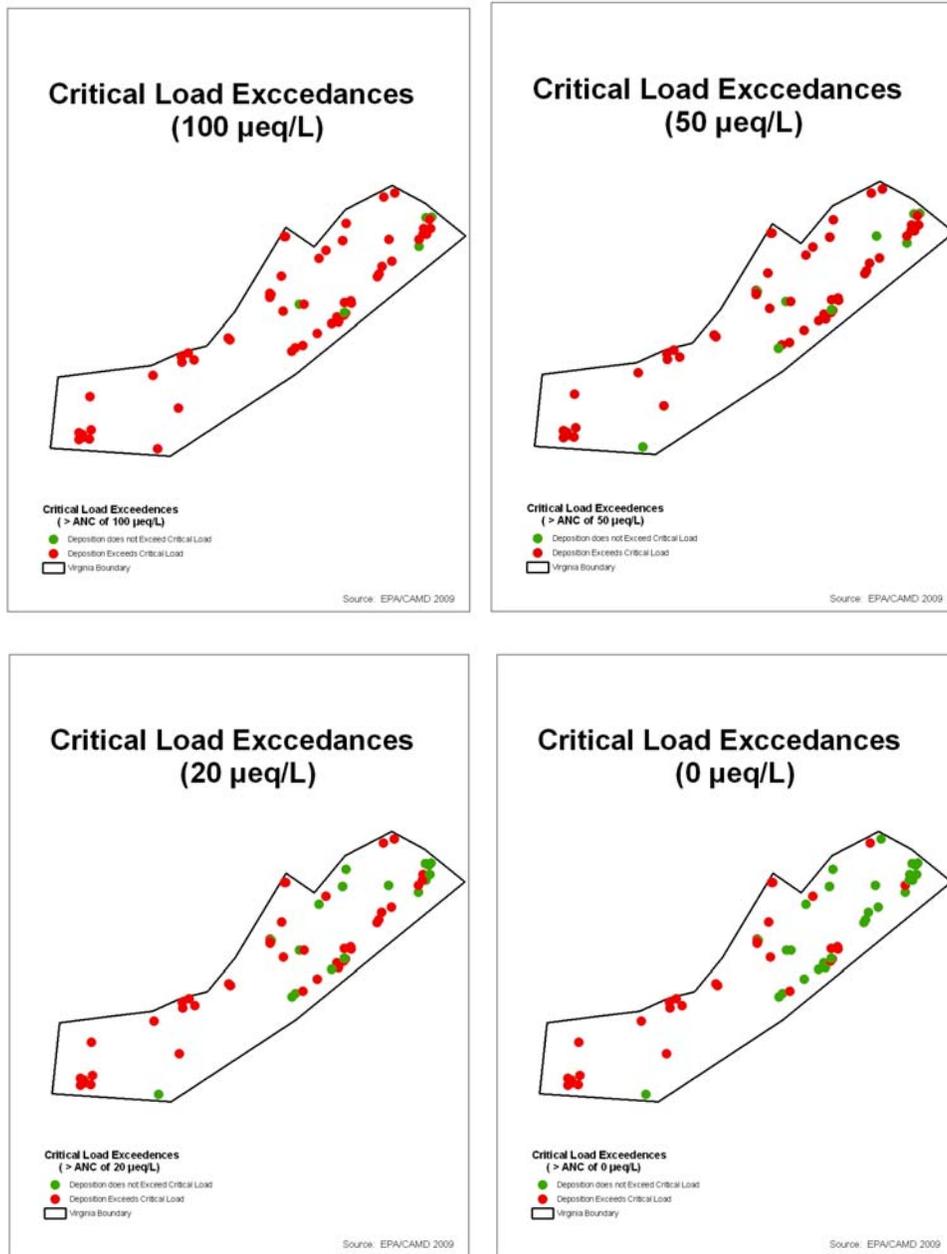


Figure 4.2-21. Critical load exceedances for acid neutralizing capacity concentrations of 0, 20, 50, and 100 $\mu\text{eq/L}$ for streams in the Shenandoah Case Study Area. Green dots represent lakes where current total nitrogen and sulfur deposition is below the critical load and that maintain an acid neutralizing capacity concentration of 0, 20, 50, and 100 $\mu\text{eq/L}$, respectively. Red dots represent streams where current total nitrogen and sulfur deposition exceeds the critical load, indicating they are currently impacted by acidifying deposition. See Table 4.2-5.

Table 4.2-5. Critical Load Exceedances (Nitrogen + Sulfur Deposition > Critical Load) for 60 Modeled Streams Within the VTSSS-LTM Monitoring Program in the Shenandoah Case Study Area. “No. Streams” Indicates the Number of Streams at the Given Acid Neutralizing Capacity Limit; “% Streams” Indicates the Total Percentage of Streams at the Given Acid Neutralizing Capacity Limit.

ANC Limit 100 µeq/L		ANC Limit 50 µeq/L		ANC Limit 20 µeq/L		ANC Limit 0 µeq/L	
No. Streams	% Streams	No. Streams	% Streams	No. Streams	% Streams	No. Streams	% Streams
55	92	51	85	43	72	31	52
Stream No. = 60							

1
2 **Recovery from acidification given current emission reductions.**
3 Based on a deposition scenario that maintains current emission levels to 2020 and 2050, a
4 large number of streams in the Shenandoah Case Study Area will still have *Elevated to Acute*
5 problems with acidity. In the short term (i.e., by the year 2020) and in the long term (i.e., by the
6 year 2050), the response of the 60 modeled streams shows no improvement in the number of
7 streams that are “not acidic.” In fact, the modeling suggests conditions may get worse by 2050
8 under current emission levels. From 2006 to 2050, the percentage of streams in *Acute Concern*
9 increases by 5%, while the percentage of streams in *Moderate Concern* decreases by 5%.

10 **4.2.5 Degree of Extrapolation to Larger Assessment Areas**

11 The EPA EMAP and Regional-EMAP (REMAP) surveys have been conducted on lakes
12 and streams throughout the country with the objective of characterizing ecological condition
13 across populations of surface waters. EMAP surveys are probability surveys where sites are
14 picked using a spatially balanced systematic randomized sample, so that the results can be used
15 to make estimates of regional extent of condition (e.g., number of lakes, length of stream).
16 Sampling typically consists of measures of aquatic biota, water chemistry, and physical habitat.
17 With respect to acidifying deposition effects, two EMAP surveys were conducted in the 1990s:
18 the Northeastern Lake Survey and the Mid-Atlantic Highlands Assessment (MAHA) of streams.
19 To make more precise estimates of the effects of acidifying deposition, the sampling grid was
20 intensified to increase the sample-site density in the Adirondack Case Study Area and New
21 England Upland areas known to be susceptible to acidifying deposition. The MAHA study was
22 conducted on 503 stream sites from 1993 to 1995 in the states of West Virginia, Virginia,

1 Pennsylvania, Maryland, and Delaware and the Catskill Mountain region of New York (Herlihy
2 et al., 2000). Results from both of these surveys were used to develop and select the sampling
3 sites for the TIME program.

4 The TIME program and the LTM program are two surface water chemistry monitoring
5 programs, administered by EPA, that inform the assessment of aquatic ecosystem responses to
6 changes in atmospheric deposition. These efforts focus on portions of the United States most
7 affected by the acidifying influence of total sulfur and nitrogen deposition, including lakes in the
8 Adirondack Case Study Area and in New England, and streams in the Shenandoah Case Study
9 Area.

10 At the core of the TIME project is the concept of probability sampling, whereby each
11 sampling site is chosen statistically from a predefined target population. The target populations
12 in these regions include lakes and streams likely to be responsive to changes in acidifying
13 deposition, defined in terms of ANC. Measurement of Gran ANC uses the Gran technique to find
14 the inflection point in an acid-base titration of a water sample (Gran, 1952). In the Northeast, the
15 TIME target population consists of lakes with a Gran ANC <100 $\mu\text{eq/L}$. In the mid-Atlantic, the
16 target population is upland streams with Gran ANC <100 $\mu\text{eq/L}$. In both regions, the sample sites
17 selected for future monitoring were selected from the EMAP survey sites in the region that met
18 the TIME target population definition. Each lake or stream is sampled annually (in summer for
19 lakes; in spring for streams), and results are extrapolated with known confidence to the target
20 population(s) as a whole using the EMAP site population expansion factors or weights (Larsen et
21 al., 1994; Larsen and Urquhart, 1993; Stoddard et al., 1996; Urquhart et al., 1998).

22 Data from 43 Adirondack Case Study Area lakes can be extrapolated to the target
23 population of low ANC lakes in that region. There are about 1,000 low-ANC Adirondack Case
24 Study Area lakes, out of a total population of 1,842 lakes with surface area greater than 1 hectare
25 (ha). Data from 30 lakes (representing about 1,500 low-ANC lakes, out of a total population of
26 6,800) form the basis for TIME monitoring in New England. Probability monitoring of mid-
27 Atlantic streams began in 1993. Stoddard et al. (2003) analyzed data from 30 low-ANC streams
28 in the Northern Appalachian Plateau (representing about 24,000 kilometer (km) of low-ANC
29 stream length out of a total stream length of 42,000 km). After pooling TIME target sites taken
30 from both MAHA and another denser random sample in 1998, there are now 21 TIME sites in
31 the Blue Ridge and Ridge and Valley that can be used for trend detection in this aggregate
32 ecoregion in the mid-Atlantic in addition to the Northern Appalachian Plateau ecoregion.

1 As a complement to the statistical lake and stream sampling in TIME, the LTM program
2 samples a subset of generally acid-sensitive lakes and streams that have long-term data, many
3 dating back to the early 1980s. These sites are sampled 3 to 15 times per year. Monitored water
4 chemistry variables include pH, ANC, major anions and cations, monomeric Al, Si, specific
5 conductance, dissolved organic carbon, and dissolved inorganic carbon. Details of LTM data
6 from each region include the following:

- 7 ▪ **New England lakes:** Data from 24 New England lakes were available for the trend
8 analysis reported by Stoddard et al. (2003) for the time period 1990 to 2000. The majority
9 of New England LTM lakes have mean Gran ANC values ranging from 20 to 100 µeq/L;
10 two higher ANC lakes (Gran ANC between 100 and 200 µeq/L) are also monitored.
- 11 ▪ **Adirondack lakes:** The trend analysis of Stoddard et al. (2003) included data from 48
12 Adirondack lakes. Sixteen of the lakes have been monitored since the early 1980s; the
13 others were added to the program in the 1990s. The Adirondack LTM dataset includes
14 both seepage and drainage lakes, most with Gran ANC values in the range –50 to 100
15 µeq/L; three lakes with Gran ANC between 100 µeq/L and 200 µeq/L are also monitored.
- 16 ▪ **Appalachian Plateau streams:** Data from four streams in the Catskill Mountains
17 (collected by the USGS; Murdoch and Stoddard, 1993) and five streams in Pennsylvania
18 (collected by Pennsylvania State University; DeWalle and Swistock, 1994) were
19 analyzed by Stoddard et al. (2003). All of the Northern Appalachian LTM streams have
20 mean Gran ANC values in the range 25 to 50 µeq/L.
- 21 ▪ **Upper Midwest lakes:** Forty lakes in the Upper Midwest were originally included in the
22 LTM project, and due to funding constraints, sampling has continued at only a subset of
23 Wisconsin lakes, as well as an independent subset of seepage lakes in the state. The data
24 reported by Stoddard et al. (2003) included 16 lakes (both drainage and seepage) sampled
25 quarterly (Webster et al., 1993) and 22 seepage lakes sampled annually in the 1990s. All
26 of the Upper Midwest LTM lakes exhibit mean Gran ANC values from 30 to 80 µeq/L.
- 27 ▪ **Ridge/Blue Ridge streams:** Data from the Ridge and Blue Ridge provinces consist of a
28 large number of streams sampled quarterly throughout the 1990s as part of the Virginia
29 Trout Stream Sensitivity Study (Webb et al., 1989) and a small number of streams
30 sampled more intensively (as in the Northern Appalachian Plateau). A total of 69 streams
31 had sufficient data for the trend analyses by Stoddard et al. (2003). All of these streams

1 were located in the Ridge section of the Ridge and Valley province or within the Blue
2 Ridge province, and all were within the state of Virginia. Mean Gran ANC values for the
3 Ridge and Blue Ridge data range from 15 to 200 $\mu\text{eq/L}$, with 7 of the 69 sites exhibiting
4 mean Gran ANC $>100 \mu\text{eq/L}$.

5 Appendix 4's Attachment 4.B of the Aquatic Acidification case study report provides a
6 more complete discussion of the EMAP/TIME/LTM programs.

7 **4.2.6 Current Conditions for the Adirondack Case Study Area and the** 8 **Shenandoah Case Study Area**

9 ***4.2.6.1 Regional Assessment of All Lakes in the Adirondack Case Study Area***

10 Estimation of the regional risk of the current the levels of NO_x and SO_2 ambient
11 concentrations and deposition onto acidification in lakes requires a scaling up of the risk derived
12 from the 169 modeled lakes to represent the risk of the entire population of lakes in the
13 Adirondack Case Study Area. One hundred 17 lakes of the 169 lakes modeled for critical loads
14 are part of a subset of 1,842 lakes in the Adirondack Case Study Area, which include all lakes
15 from 0.5 to 2,000 ha in size and at least 1 m in depth. Using weighting factors derived from the
16 EMAP probability survey and critical load calculations from the 117 lakes, estimates of
17 exceedances were derived for the entire 1,842 lakes in the Adirondack Case Study Area. Based
18 on this approach, 945, 666, 242, and 135 lakes exceed their critical load for ANC limits of 100,
19 50, 20, and 0 $\mu\text{eq/L}$, respectively (**Table 4.2-6**).

20 Given a low level of protection from acidification (i.e., an ANC limit of 20 $\mu\text{eq/L}$), the
21 current risk of acidification is 242 lakes or 13%. Because some lakes in the Adirondack Case
22 Study Area have natural sources of acidity, some lakes would have never had ANC
23 concentrations of above 50 and 100 $\mu\text{eq/L}$. For this reason, the actual number of lakes at risk of
24 acidification at an ANC level of 50 and 100 $\mu\text{eq/L}$ is lower than the estimate based on the critical
25 load alone. Using the hindcast simulation from the MAGIC model, 11% of modeled lakes have
26 natural ANC concentrations of less than 50 $\mu\text{eq/L}$. Excluding these naturally acidic lakes, the
27 current risk of acidification is 666 lakes or 36% for a moderate protective ANC concentration of
28 50 $\mu\text{eq/L}$. For an ANC level of 100 $\mu\text{eq/L}$, 51% of lakes have natural ANC concentrations below
29 100 $\mu\text{eq/L}$. Excluding these naturally acidic lakes, the current risk is 945 lakes or 51% for a
30 protective ANC concentration of 100 $\mu\text{eq/L}$. Even with corrections for natural acidity, 8 to 41%

1 of lakes in the Adirondack Case Study Area are at risk of acidification given current ambient
2 concentration of NO_x and SO₂.

3 Because some lakes in the Adirondack Case Study Area have natural sources of acidity,
4 some lakes would never have ANC concentrations above 50 or 100 µeq/L, even in the absence of
5 all anthropogenically derived acidifying deposition. Based on the hindcast simulations of 44
6 lakes using the MAGIC model, no modeled lakes have ANC levels below 20 µeq/L. However, 5
7 modeled lakes or 11% have ANC concentrations between 22 and 47 µeq/L. This equates to
8 approximately 300 lakes or 16% of the representative population of lakes in the Adirondack
9 Case Study Area that likely had preacidification ANC concentrations below 50 µeq/L. On the
10 other hand, potentially more than 52% of lakes likely had preacidification ANC concentrations
11 below 100 µeq/L. The higher percentage of lakes in the regional population compared to the
12 modeled population is because the lake classes or sizes likely to have a preacidification ANC
13 concentration below 50 or 100 µeq/L are more abundant in the Adirondack Case Study Area than
14 lakes with a preacidification ANC concentration above 50 or 100 µeq/L.

Table 4.2-6. Critical Load Exceedances (Nitrogen + Sulfur Deposition > Critical Load) for the Regional Population of 1,849 Lakes in the Adirondack Case Study Area That Are from 0.5 to 2000 ha in Size and at Least 1 m in Depth. Estimates Are Based on the EMAP Lake Probability Survey of 1991 to 1994.

ANC Limit 100 µeq/L		ANC Limit 50 µeq/L		ANC Limit 20 µeq/L		ANC Limit 0 µeq/L	
No. Lakes	% Lakes	No. Lakes	% Lakes	No. Lakes	% Lakes	No. Lakes	% Lakes
945	51	666	36	242	13	135	7
Lake No. = 1842							

15

16 **4.2.6.2 Regional Assessment of All Streams in the Shenandoah Case Study Area**

17 The 60 trout streams modeled are characteristic of first- and second-order streams on
18 nonlimestone bedrock in the Shenandoah Case Study Area. Because of the strong relationship
19 between bedrock geology and ANC in this region, it is possible to consider the results in the
20 context of similar trout streams in the Southern Appalachian Mountains that have the same
21 bedrock geology and size. The total number of brook trout streams in the Shenandoah Case
22 Study Area represented is 440, of which 308 lie on limestone and/or have not been significantly
23 affected by human activity within their watersheds. In addition, the 60 modeled streams are a

1 subset of 344 streams sampled by the Virginia Trout Stream Sensitivity Study, of which 304
 2 represent the different sizes and bedrock types found to be sensitive to acidification. Using the
 3 304 streams to which the analysis applies directly as the total, 279, 258, 218, and 157 streams
 4 exceed their critical load for 2002 deposition with critical limits of 100, 50, 20, and 0 $\mu\text{eq/L}$,
 5 respectively. However, it is likely that many more of the ~12,000 trout streams in the
 6 Shenandoah Case Study Area would exceed their critical load given the extent of similar bedrock
 7 geology outside the study area in the Southern Appalachian Mountains.

8 **4.2.7 Ecological Effect Function for Aquatic Acidification**

9 Atmospheric deposition of NO_x and SO_x contributes to acidification in aquatic
 10 ecosystems through the input of acid anions, such as NO_3^- and SO_4^{2-} . The acid balance of
 11 headwater lakes and streams is controlled by the level of this acidifying deposition of NO_3^- and
 12 SO_4^{2-} and a host of catchment processes and environmental factors that affect the level of base
 13 cations (e.g., Ca^+ , Mg^+) concentrations and the sinks of nitrogen and sulfur in the lake and
 14 terrestrial catchment. The biotic integrity of freshwater ecosystems is then a function of the acid-
 15 base balance and the resulting acidity-related stress on the biota that occupy the water.

16 The calculated ANC of the surface waters is a measure of the acid-base balance:

$$17 \quad \text{ANC} = [\text{BC}]^* - [\text{AN}]^* \quad (1)$$

18 where $[\text{BC}]^*$ and $[\text{AN}]^*$ are the sum of base cations and acid anions (NO_3^- and SO_4^-),
 19 respectively, in the surface water accounting for the effects input of Cl^- . Although ANC does not
 20 directly affect the health of biotic communities, it ameliorates acidity-related biotic stress that
 21 provides an “ecological indicator” of overall integrity of the ecosystem.

22 The ANC concentration then provides a link between the surface water acidification and
 23 the ecological integrity of the aquatic community where a given level of ANC corresponds to an
 24 ecological effect (Table 4.2-1). It also provides a link between the deposition of NO_x and SO_x
 25 and the acidification through the input of acid anions of NO_3^- and SO_4^- .

26 Equation (1) forms the basis of the linkage between deposition and surface water acidic
 27 condition and the modeling approach used. Given some “target” ANC concentration $[\text{ANC}_{\text{limit}}]$,
 28 which protects biological integrity, the amount of deposition of acid anions (AN) or depositional
 29 load ($\text{DL}(\text{N}) + \text{DL}(\text{S})$) is simply the input flux of acid anions from atmospheric deposition that
 30 result in a surface water ANC concentration equal to the $[\text{ANC}_{\text{limit}}]$ when balanced by the

1 sustainable flux of base cations input and the sinks of nitrogen and sulfur in the lake and
 2 watershed catchment. The sustainable flux of base cations input and sinks of nitrogen and sulfur
 3 is equal to the uptake (N_{upt}), immobilization (N_{imm}), and denitrification (N_{den}) of nitrogen in the
 4 catchment, the in-lake retention of nitrogen (N_{ret}) and sulfur (S_{ret}), and the preindustrial flux of
 5 base cations ($[BC]_0^*$) to the watershed. Thus, the amount of deposition that will maintain an
 6 ANC level above an ANC_{limit} is described as

$$7 \quad DL(N) + DL(S) = \{fN_{\text{upt}} + (1 - r)(N_{\text{imm}} + N_{\text{den}}) + (N_{\text{ret}} + S_{\text{ret}})\} + ([BC]_0^* - [ANC_{\text{limit}}])Q \quad (2)$$

8 where f and r are dimensionless parameters that define the fraction of forest cover in the
 9 catchment and the lake/catchment ratio, respectively, and Q is runoff. To convert surface water
 10 concentrations into surface water fluxes, multiply by runoff (Q) (in m/yr) from the site. Several
 11 major assumptions are made: (1) steady-state conditions exist, (2) the effect of nutrient cycling
 12 between plants and soil is ignored, (3) there are no significant nitrogen inputs from sources other
 13 than atmospheric deposition, (4) ammonium leaching is negligible because any inputs are either
 14 taken up by biota or adsorbed onto soils or nitrate compounds, and (5) long-term sinks of sulfate
 15 in the catchment soils are negligible.

16 It is not possible to define a maximal loading for a single total of acidity (i.e., both
 17 nitrogen and sulfur deposition) because the acid anions sulfate and nitrate behave differently in
 18 the way they are transported with hydrogen ions; one unit of deposition of sulfur will not have
 19 the same net effect on surface water ANC as an equivalent unit of nitrogen deposition. However,
 20 the individual maximum and minimum depositional loads for nitrogen and sulfur are defined
 21 when nitrogen or sulfur do not contribute to the acidity in the water. The maximum depositional
 22 load for sulfur ($DL_{\text{max}}(S)$) is equal to the amount of sulfur the catchment can remove and still
 23 maintain an ANC concentration above the ANC_{limit} :

$$24 \quad DL_{\text{max}}(S) = [([BC]_0^* - [ANC_{\text{level}}])Q] / (1 - p_s) \quad (3)$$

25 when nitrogen deposition does not contribute to the acidity balance and where p_s defines the
 26 fraction of in-lake retention of S_{ret} . Given the assumption that the long-term sinks of sulfate in the
 27 catchment soils are negligible, the amount of sulfur entering the catchment is equal to the amount
 28 loaded to the surface water. For this reason, the minimal amount of sulfur is equal to zero:

$$29 \quad DL_{\text{min}}(S) = 0 \quad (4)$$

1 In the case of nitrogen, $DL_{\min}(N)$ is the minimum amount of deposition of total nitrogen ($NH_x +$
 2 NO_x) that catchment processes can effectively remove (e.g., $N_{\text{upt}} + N_{\text{imm}} + N_{\text{den}} + N_{\text{ret}}$) without
 3 contributing to the acidic balance:

$$4 \quad DL_{\min}(N) = fN_{\text{upt}} + (1-r)(N_{\text{imm}} + N_{\text{den}}) \quad (5)$$

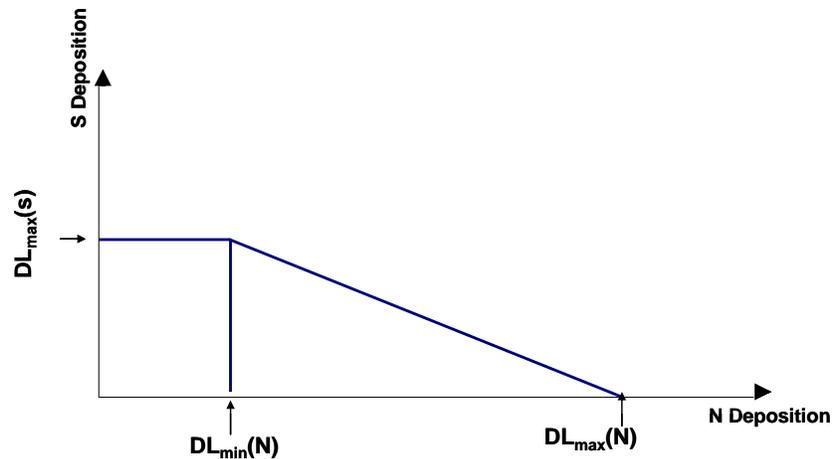
5 The $DL_{\max}(N)$ is the load for total nitrogen deposition when sulfur deposition is equal to
 6 zero:

$$7 \quad DL_{\max}(N) = fN_{\text{upt}} + (1-r)(N_{\text{imm}} + N_{\text{den}}) + \left[\frac{([BC]_0^* - [ANC_{\text{level}}])Q}{(1-p_n)} \right] \quad (6)$$

8 where p_n defines the fraction of in-lake retention of N_{ret} .

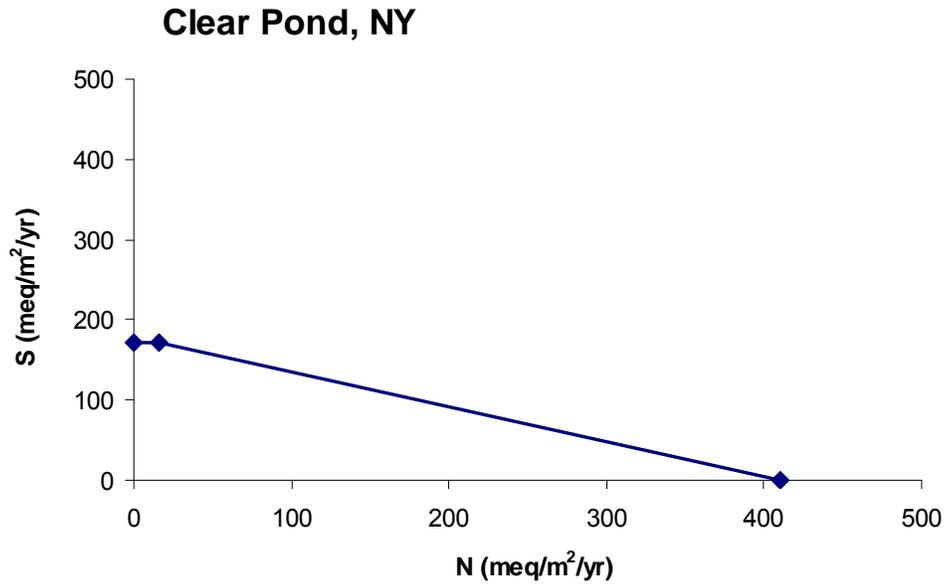
9 In reality, neither nitrogen nor sulfur deposition will ever be zero, so the depositional load
 10 for the deposition of one is fixed by the deposition of the other, according to the line defining in

11 **Figure 4.2-22.**

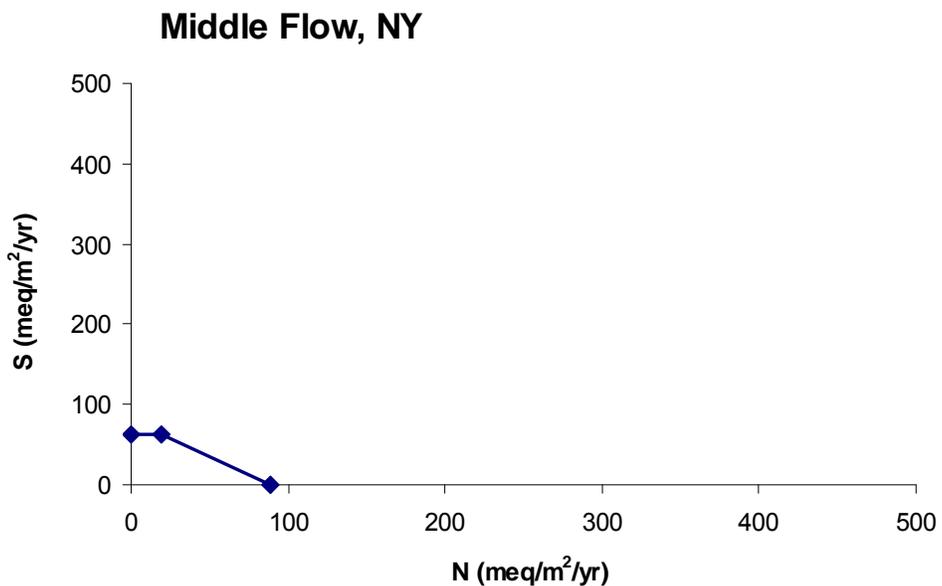


12
 13 **Figure 4.2-22.** The depositional load function defined by the model.

14 The thick lines indicate all possible pairs of depositional loads of nitrogen and sulfur
 15 acidity that a catchment can receive and still maintain an ANC concentration equal to its
 16 ANClimit. Note that in the above formulation, individual depositional loads of nitrogen and
 17 sulfur are not specified; each pair of depositions (S_{dep} and N_{dep}) fulfilling Equations 2 through 6.
 18 **(Figure 4.2-23)** shows the depositional load function for two lakes in New York



1



2

3 **Figure 4.2-23** Deposition load graphs for Clear Pond and Middle Flow Lake, New York.4 **4.2.8 Uncertainty and Variability**

5 Uncertainty was examined in both the MAGIC-derived values of surface water chemistry
 6 and critical load estimates from the SSWC model. In both cases, uncertainty surrounds the
 7 parameters that are used in the model calibrations. For example, the strength of the critical load
 8 estimate calculations relies on the ability to estimate the catchment-average base cation supply

1 (i.e., input of base cations from weathering of bedrock and soils and air). The ability to
2 accurately estimate the catchment supply of base cations to a water body is still poorly known for
3 the United States. This is important because the catchment supply of base cations from the
4 weathering of bedrock and soils is the factor that has the most influence on the critical load
5 calculation and has the largest uncertainty (Li and McNulty, 2007).

6 Although the F-factor approach and SSWC model have been widely published and
7 analyzed in Canada and Europe, and have been applied in the United States (e.g., Dupont et al.,
8 2005), their utility in critical load calculations is still unclear. For this reason, an uncertainty
9 analysis of the SSWC critical load model was completed to evaluate the uncertainty in the
10 modeling parameters. A probabilistic analysis using a range of parameter uncertainties was used.
11 The probabilistic framework is Monte Carlo, whereby each SSWC input parameter varies
12 according to specified probability distributions. Within Monte Carlo analysis, models are run a
13 sufficient number of times (i.e., 2,000 times) to capture the range of behaviors represented by all
14 variable inputs to the SSWC model (see model description). In this case study, multiple values
15 were selected for several parameters in the SSWC calculations, based on published values and
16 regional environmental constraints. The analysis tabulated the number of lakes where the
17 confidence interval is entirely below the critical load, the confidence interval is entirely above
18 the critical load, and the confidence interval straddles zero. Similar results are given for the
19 number of lakes with all realizations above the critical load, all realizations below the critical
20 load, and some realizations above and some below the critical load.

21 To evaluate the degree to which critical load estimates could change with a range of
22 possible parameter values, a simple summary of the Monte Carlo analysis was completed to
23 determine the critical load amount (meq/m²/yr) and the percentage change, using a levels of
24 protection of ANC = 50 µg/L. The comparisons of critical loads revealed that changes in critical
25 load values could range from 3 to 34 meq/m²/yr, depending on the magnitude of the critical load
26 itself. This corresponded to percent differences ranging on average from 0% to 8%, with a few
27 values exceeding 10%. Using the 95% confidence interval, the percent difference in the number
28 of waterbodies exceeding their critical load compared to the mean percentage was calculated.
29 The comparisons of critical load exceedance rates revealed an average difference of 5% (range 2
30 to 8%), meaning the percent of waterbodies that exceed their critical load varied by about 5%.
31 The results suggest a relatively robust estimate of critical loads and exceedance rates for the case
32 study areas. This analysis may understate the actual uncertainty because some of the range and

1 distribution types of parameters in the SSWC model are not well known for the United States at
2 this time.

3 The uncertainty in the water quality estimates (i.e., ANC) from MAGIC was derived by
4 running multiple calibrations. These simulation uncertainty estimates were derived from the
5 multiple calibrations at each site provided by the “fuzzy optimization” procedure employed in
6 this project. For each of the modeled sites, 10 distinct calibrations were performed with the target
7 values, parameter values, and deposition inputs for each calibration, reflecting the uncertainty
8 inherent in the observed data for the individual site. The effects of the uncertainty in the
9 assumptions made in calibrating the model (and the inherent uncertainties in the data available)
10 can be assessed by using all successful calibrations for a site when simulating the response to
11 different scenarios of future deposition. The model then produces an ensemble of simulated
12 values for each site, e.g., a median ANC.

13 Based on the MAGIC model simulations, the 95% confidence interval for the pre-
14 acidification and current average ANC concentrations of 44 modeled lakes is 106.8 to 134.0 and
15 50.5 to 81.8 $\mu\text{eq/L}$, respectively, which is on average a 15 $\mu\text{eq/L}$ difference in ANC
16 concentrations or 10 percent. The 95% confidence interval for pre-acidification and current
17 average ANC concentrations of the 60 modeled streams is 91.9 to 110.9 and 53.4 to 62.4 $\mu\text{eq/L}$,
18 respectively, which is on average 8 $\mu\text{eq/L}$ difference in ANC concentration or 5 percent.

19 **4.3 TERRESTRIAL ACIDIFICATION**

20 **4.3.1 Ecological Indicators, Ecological Responses, and Ecosystem Services**

21 **4.3.1.1 Ecological Indicators**

22 The ISA (U.S. EPA, 2008) identified a variety
23 of indicators supported by the literature that can be used
24 to measure the effects of acidification in soils. Much of
25 the literature discussing terrestrial acidification focuses on Ca^{2+} and Al as the primary indicators
26 of detrimental effects for trees and other terrestrial vegetation. Both of these indicators are
27 strongly influenced by soil acidification, and both have been shown to have quantitative links to
28 tree health (see Appendix 5 for more information).

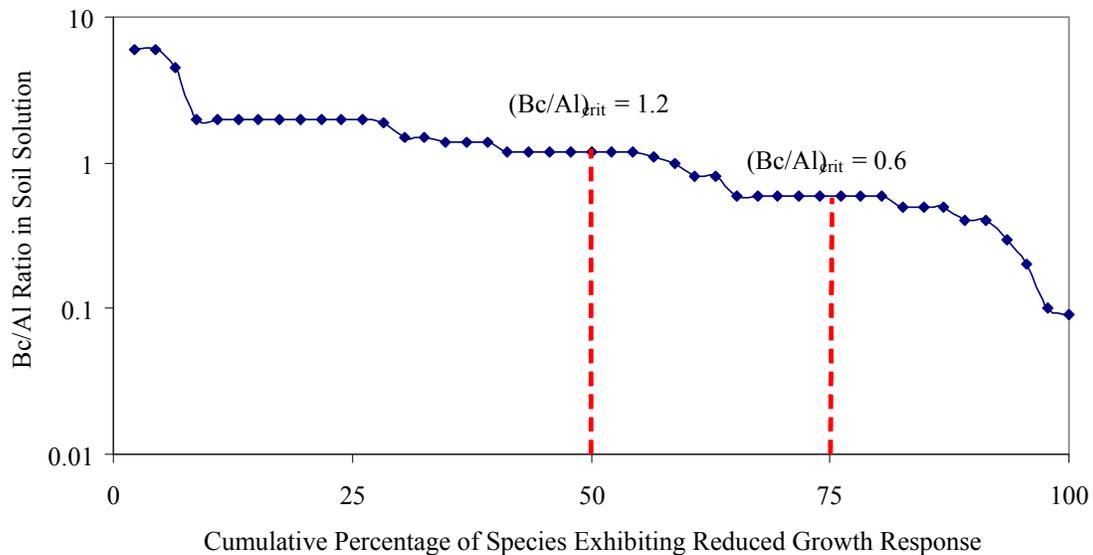
Indicator: The Bc/Al ratio in the soils solution was selected as the indicator to estimate critical deposition loads of acidity for the Terrestrial Acidification Case Study.

29 Therefore, the Ca/Al ratio in soil solution was selected as the basis for the indicator in the
30 Terrestrial Acidification Case Study (Appendix 5) to evaluate the critical load of acidity in

1 terrestrial systems. Within the calculations of critical loads, the base cation (Bc) to Al ratio
 2 (Bc/Al) was used to represent the Ca/Al indicator. This approach was selected because the Bc
 3 variable consists of multiple base cations (Ca^{2+} , Mg^{2+} , and K^+), and the Bc/Al ratio is the most
 4 commonly used indicator or critical ratio ($\text{Bc/Al}_{\text{crit}}$) in estimations of acid load (McNulty et al.,
 5 2007; Ouimet et al., 2006; UNECE, 2004).

6 **4.3.1.2 Ecological Responses**

7 In a meta-analysis of studies that explored the relationship between Bc/Al ratio in soil
 8 solution and tree growth, Sverdrup and Warfvinge (1993) reported the Bc/Al ratios at which
 9 growth was reduced by 20% relative to control trees. **Figure 4.3-1** presents the findings of
 10 Sverdrup and Warfvinge (1993) based on 46 of the tree species that grow in North America. This
 11 summary indicates that there is a 50% chance of negative tree response (i.e., >20% reduced
 12 growth) at a soil solution Bc/Al ratio of 1.2 and a 75% chance at a Bc/Al ratio of 0.6. These
 13 findings clearly demonstrate a relationship between Bc/Al ratio and tree health; as the Bc/Al is
 14 reduced, there is a greater likelihood of a negative impact on tree health.



15

16

17

18

19

Figure 4.3-1. The relationship between the Bc/Al ratio in soil solution and the percentage of tree species (found growing in North America) exhibiting a 20% reduction in growth relative to controls (after Sverdrup and Warfvinge, 1993).

20

21

22

The tree species most commonly studied in North America to assess the impacts of acidification due to total nitrogen and sulfur deposition are red spruce (i.e., *Picea Rubens*, a coniferous tree species) and sugar maple (i.e., *Acer saccharum*, a deciduous tree species). At a

1 Bc/Al soil solution ratio of approximately 1.2, red spruce growth can be reduced by 20%, and a
2 similar reduction in growth may be experienced by sugar maple at a Bc/Al ratio of 0.6 (Sverdrup
3 and Warfvinge 1993). Both species are also found in the eastern United States, and soil
4 acidification is widespread throughout this area (Warby et al., 2009).

5 Red spruce is found scattered throughout high-elevation sites in the Appalachian
6 Mountains, including the southern peaks. Noticeable fractions of the canopy red spruce died
7 within the Adirondack, Green, and White mountains in the 1970s and 1980s. Acidifying
8 deposition has been implicated in this decline because of links between tree stress from Al
9 toxicity and increased freezing injury (DeHayes et al., 1999). Within the southeastern United
10 States, periods of red spruce growth decline slowed after the 1980s, when a corresponding
11 decrease in SO₂ emissions was recorded in the United States (Webster et al., 2004). Red spruce
12 has been shown to have an increased instance of foliar winter injury and bud mortality due to
13 imbalanced Al and Ca²⁺ levels in soils at locations in Vermont and surrounding states. A
14 decrease in cold and winter weather tolerance leads to an increase in freezing injuries to red
15 spruce, placing the species at a greater chance of declining overall forest health. Soil nutrient
16 imbalances and deficiencies can reduce the ability of a tree to respond to stresses, such as insect
17 defoliation, drought, and cold weather damage (DeHayes et al., 1999; Driscoll et al., 2001).
18 Based on the research conducted to date, important factors related to the high mortality rates and
19 decreased growth trends of red spruce include depletion of base cations in upper soil horizons by
20 acidifying deposition, Al toxicity to tree roots, and accelerated leaching of base cations from
21 foliage as a consequence of acidifying deposition (U.S. EPA, 2008, Section 3.2.2.3). Additional
22 linkages between acidifying deposition and red spruce physiological responses are indicated in
23 **Table 4.3-1.**

24 Sugar maple is found throughout the northeastern United States and the central
25 Appalachian Mountain region. This species has been declining in the eastern United States since
26 the 1950s. Studies on sugar maple have found that this decline in growth is related to both
27 acidifying deposition and base-poor soils on geologies dominated by sandstone or other base-
28 poor substrates (Bailey et al., 2004; Horsley et al., 2000). These site conditions are representative
29 of the conditions expected to be most susceptible to impacts of acidifying deposition because of
30 probable low initial base cation pools and high base cation leaching losses (U.S. EPA, 2008,
31 Section 3.2.2.3). The probability of a decrease in crown vigor or an increase in tree mortality has
32 been noted to increase at sites with low Ca²⁺ and Mg²⁺ as a result of leaching caused by

1 acidifying deposition (Drohan and Sharpe, 1997). Low levels of Ca^{2+} in leaves and soils have
 2 been shown to be related to lower rates of photosynthesis and higher antioxidant enzyme activity
 3 in sugar maple stands in Pennsylvania (St. Clair et al., 2005). Additionally, plots of sugar maples
 4 in decline were found to have Ca^{2+}/Al ratios less than 1, as well as lower base cation
 5 concentrations and pH values compared with plots of healthy sugar maples (Drohan et al., 2002).
 6 These indicators have all been shown to be related to the deposition of atmospheric nitrogen and
 7 sulfur. Additional linkages between acidifying deposition and sugar maple physiological
 8 responses are indicated in **Table 4.3-1**.

Table 4.3-1. Summary of Linkages Between Acidifying Deposition, Biogeochemical Processes That Affect Ca^{2+} , Physiological Processes That Are Influenced by Ca^{2+} , and Effect on Forest Function

Biogeochemical Response to Acidifying deposition	Physiological Response	Effect on Forest Function
Leach Ca^{2+} from leaf membrane	Decrease the cold tolerance of needles in red spruce	Loss of current-year needles in red spruce
Reduce the ratio of Ca^{2+}/Al in soil and soil solutions	Dysfunction in fine roots of red spruce blocks uptake of Ca^{2+}	Decreased growth and increased susceptibility to stress in red spruce
Reduce the ratio of Ca^{2+}/Al in soil and soil solutions	More energy is used to acquire Ca^{2+} in soils with low Ca^{2+}/Al ratios	Decreased growth and increased photosynthetic allocation to red spruce roots
Reduce the availability of nutrient cations in marginal soils	Sugar maples on drought-prone or nutrient-poor soils are less able to withstand stresses	Episodic dieback and growth impairment in sugar maple
Source: Fenn et al., 2006.		

9
 10 In summary, the acidification of soils negatively
 11 impacts the health, growth, and vigor of red spruce and
 12 sugar maple. Mortality and susceptibility to disease and
 13 injury can be increased and growth decreased with
 14 acidifying deposition. Therefore, the health of sugar maple and red spruce was used as the
 15 endpoints (ecological responses) to evaluate acidification in terrestrial systems.

End Point: The health of sugar maple and red spruce was selected as the endpoints to estimate critical deposition loads of acidity in this case study.

16 **4.3.1.3 Ecosystem Services**

17 A number of impacts on the ecological endpoints of forest health, water quality, and
 18 habitat exist, including the following:

- 1 ▪ Decline in habitat for threatened and endangered species—cultural
- 2 ▪ Decline in forest aesthetics—cultural
- 3 ▪ Decline in forest productivity—provisioning
- 4 ▪ Increases in forest soil erosion and reductions in water retention—cultural and regulating.

5 These impacts are described below.

6 (Existing ecosystem services that are primarily impacted by the terrestrial acidification
7 resulting from total nitrogen and sulfur deposition are being quantified for the Risk and Exposure
8 Assessment.)

9 **Provisioning Services.** Forests in the northeastern United States provide several
10 important and valuable provisioning services, which are reflected in measures of production and
11 sales of tree products.

12 Sugar maples (also referred to as hard maples) are a particularly important commercial
13 hardwood tree species in the United States. The two main types of products derived from sugar
14 maples are wood products and maple syrup. The wood from sugar maple trees is particularly
15 hard, and its primary uses include construction, furniture, and flooring (Luzadis and Gossett,
16 1996). According to data from the U.S. Forest Service’s National Forest Inventory and Analysis
17 (FIA) database, the total removal of sugar maple saw timber from timberland in the United States
18 was almost 900 million board feet in 2006 (USFS, 2006). During winter and early spring
19 (depending, in part, on location and diurnal temperature differences), sugar maple trees also
20 generate sap that is used to produce maple syrup. From 2005 to 2007, annual production of
21 maple syrup in the United States varied between 1.2 million and 1.4 million gallons, which
22 accounted for roughly 19% of worldwide production. The total annual value of U.S. production
23 in these years varied between \$157 million and \$168 million (NASS, 2008).

24 Red spruce is a common commercial softwood species whose wood is used in a variety
25 of products including lumber, pulpwood, poles, plywood, and musical instruments. According to
26 FIA data, the total removal of red spruce saw timber from timberland in the United States was
27 328 million board feet in 2006 (USFS, 2006).

28 **Figure 4.3-2** shows and compares the value of annual production of sugar maple and red
29 spruce wood products and of maple syrup in 2006. Across states in the northeastern United
30 States, wood from sugar maple harvests consistently generated the highest total sales value of the
31 three products. Although total sales of red spruce saw timber and maple syrup were of roughly

1 the same magnitude in the United States as a whole, the red spruce harvest was concentrated in
2 Maine, whereas maple syrup production was largest in Vermont and New York.

3 **Cultural Services.** Forests in the northeastern United States are also an important source
4 of cultural ecosystem services—nonuse (i.e., existence value for threatened and endangered
5 species), recreational, and aesthetic services. Red spruce forests are home to two federally listed
6 species and one delisted species:

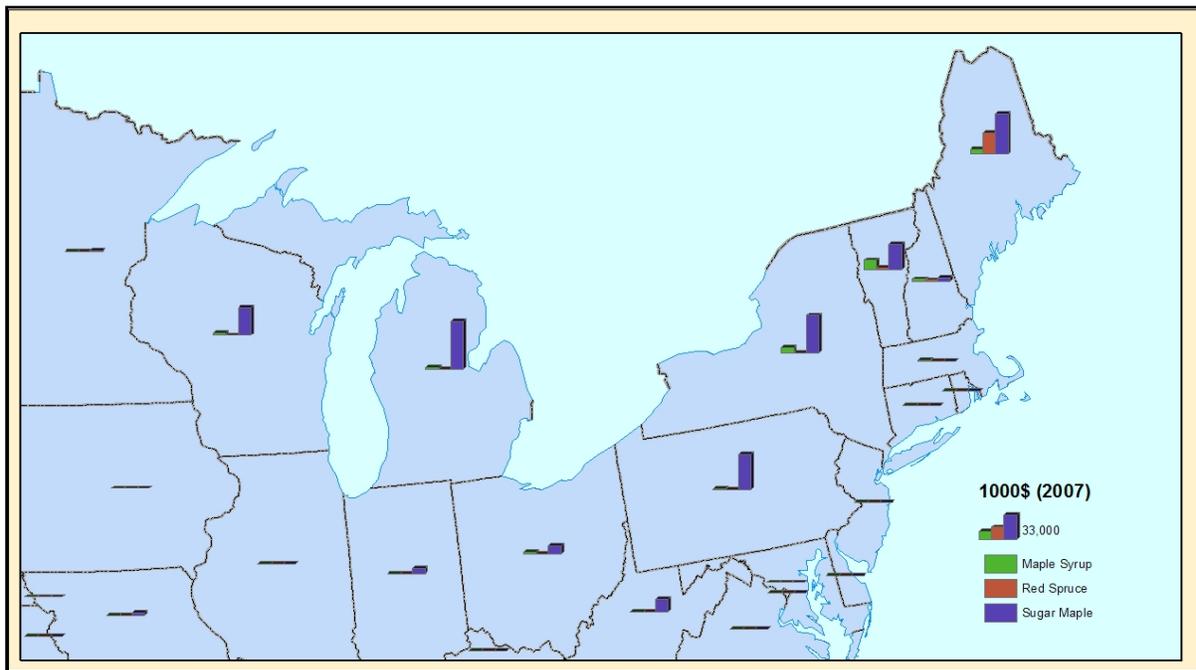
- 7 ■ Spruce-fir moss spider (*Microhexura montivaga*)—endangered
- 8 ■ Rock gnome lichen (*Gymnoderma lineare*)—endangered
- 9 ■ Virginia northern flying squirrel (*Glaucomys sabrinus fuscus*)—delisted, but important.

10 Forest lands support a wide variety of outdoor recreational activities, including fishing,
11 hiking, camping, off-road driving, hunting, and wildlife viewing. Regional statistics on
12 recreational activities that are specifically forest based are not available; however, more general
13 data on outdoor recreation provide some insights into the overall level of recreational services
14 provided by forests. For example, most recent data from the National Survey on Recreation and
15 the Environment (NSRE) indicate that, from 2004 to 2007, 31% of the U.S. adult (16 and older)
16 population visited a wilderness or primitive area during the previous year, and 32% engaged in
17 day hiking (Cordell et al., 2008). From 1999 to 2004, 16% of adults in the northeastern United
18 States¹ participated in off-road vehicle recreation, for an average of 27 days per year (Cordell
19 et al., 2005). Using the meta-analysis results reported by Kaval and Loomis (2003), which found
20 that the average consumer surplus value per day of off-road driving in the United States was
21 \$25.25 (in 2007 dollars), the implied total annual value of off-road driving recreation in the
22 northeastern United States was more than \$9.25 billion.

23 State-level data on other outdoor recreational activities associated with forests are also
24 available from the *2006 National Survey of Fishing, Hunting, and Wildlife-Associated*
25 *Recreation* (U.S. FWS and U.S. Census Bureau, 2007). Five and one-half percent of adults in the
26 northeastern United States participated in hunting, and the total number of hunting days
27 occurring in those states was 83.8 million. Data from the survey also indicated that 10% of adults
28 in northeastern states participated in wildlife viewing away from home. The total number of

¹ This area includes Connecticut, Delaware, District of Columbia, Illinois, Indiana, Maine, Maryland, Massachusetts, Michigan, New Hampshire, New Jersey, New York, Ohio, Pennsylvania, Rhode Island, Vermont, West Virginia, and Wisconsin.

1 away-from-home wildlife viewing days occurring in those states was 122.2 million in 2006. For
 2 these recreational activities in the northeastern United States, Kaval and Loomis (2003)
 3 estimated average consumer surplus values per day of \$52.36 for hunting and \$34.46 for wildlife
 4 viewing (in 2007 dollars). The implied total annual value of hunting and wildlife viewing in the
 5 northeastern United States was, therefore, \$4.38 billion and \$4.21 billion, respectively, in 2006.



6
 7 **Figure 4.3-2.** 2006 annual value of sugar maple and red spruce harvests and maple
 8 syrup production, by state.

9 As previously mentioned, it is difficult to estimate the portion of these recreational
 10 services that are specifically attributable to forests and to the health of specific tree species.
 11 However, one recreational activity that is directly dependent on forest conditions is fall color
 12 viewing. Sugar maple trees, in particular, are known for their bright colors and are, therefore, an
 13 essential aesthetic component of most fall color landscapes. Statistics on fall color viewing are
 14 much less available than for the other recreational and tourism activities; however, a few studies
 15 have documented the extent and significance of this activity. For example, based on a 1996 to
 16 1998 telephone survey of residents in the Great Lakes area, Spencer and Holecek (2007) found
 17 that roughly 30% of residents reported at least one trip in the previous year involving fall color
 18 viewing. In a separate study conducted in Vermont, Brown (2002) reported that more than 22%
 19 of households visiting Vermont in 2001 made the trip primarily for the purpose of viewing fall

1 colors. Unfortunately, data on the total number or value of these trips are not available, although
2 the high rates of participation suggest that numbers might be similar to the wildlife viewing
3 estimates reported above.

4 Although these statistics provide useful indicators of the total recreational and aesthetic
5 services derived from forests in the northeastern United States, they do not provide estimates of
6 how these services are affected by terrestrial and forest acidification. Very few empirical studies
7 have directly addressed this issue; however, there are two studies that have estimated values for
8 protecting high-elevation spruce forests in the southern Appalachian Mountains. Kramer et al.,
9 (2003) conducted a contingent valuation study estimating households' willingness to pay (WTP)
10 for programs to protect remaining high-elevation spruce forests from damages associated with air
11 pollution and insect infestation (Haefele et al., 1991; Holmes and Kramer, 1995). The study
12 collected data from 486 households using a mail survey of residents living within 500 miles of
13 Asheville, NC. The survey presented respondents with photographs representing three stages of
14 forest decline and explained that, without forest protection programs, high-elevation spruce
15 forests would all decline to worst conditions (with severe tree mortality). The survey then
16 presented two potential forest protection programs, one of which would prevent further decline
17 in forests along roads and trail corridors (one-third of the at-risk ecosystem) and the other would
18 prevent decline in all at-risk forests. Both programs would be funded by tax payments going to a
19 conservation fund. Median household WTP was estimated to be roughly \$29 (in 2007 dollars)
20 for the first program, and \$44 for the more extensive program.

21 Jenkins et al. (2002) conducted a very similar study in 1995, using a mail survey of
22 households in seven Southern Appalachian states. In this study, respondents were presented with
23 one potential program, which would maintain forest conditions at initial (status quo) levels. It
24 was explained that, without the program, forest conditions would decline to worst conditions
25 (with 75% dead trees). In contrast to the previously described study, in this survey the *initial*
26 level of forest condition was varied across respondent. In one version of the survey, the initial
27 condition was described and shown as 5% dead trees, while the other version described and
28 showed 30% dead trees. Household WTP was elicited from 232 respondents using a
29 dichotomous choice and tax payment format. The overall mean annual WTP for the forest
30 protection programs was \$208 (in 2007 dollars), which is considerably larger than the WTP
31 estimates reported by Kramer et al. (2003). One possible reason for this difference is that
32 respondents to the Jenkins et al. (2002) survey, on average, lived much closer to the affected

1 ecosystem. Multiplying the average WTP estimate from this study by the total number of
2 households in the seven-state Appalachian region results in an aggregate annual value of \$3.4
3 billion for avoiding a significant decline in the health of high-elevation spruce forests in the
4 Southern Appalachian region.

5 **Regulating Services.** Forests in the northeastern United States also support and provide a
6 wide variety of valuable regulating services, including soil stabilization and erosion control,
7 water regulation, and climate regulation (Krieger, 2001). Forest vegetation plays an important
8 role in maintaining soils in order to reduce erosion, runoff, and sedimentation that can negatively
9 impact surface waters. In addition to protecting the *quality* of water in this way, forests also help
10 store and regulate the *quantity* and flows of water in watersheds. Finally, forests help regulate
11 climate locally by trapping moisture and globally by sequestering carbon. The total value of
12 these ecosystem services is very difficult to quantify in a meaningful way, as is the reduction in
13 the value of these services associated with total nitrogen and sulfur deposition. As terrestrial
14 acidification contributes to root damages, reduced biomass growth, and tree mortality, all of
15 these services are likely to be affected; however, the magnitude of these impacts is currently very
16 uncertain.

17 **4.3.2 Characteristics of Sensitive Areas**

18 In general, forest ecosystems of the Adirondack Mountains of New York, Green
19 Mountains of Vermont, White Mountains of New Hampshire, the Allegheny Plateau of
20 Pennsylvania, and high-elevation forests in the southern Appalachian Mountains are considered
21 to be the regions most sensitive to terrestrial acidification effects from acidifying deposition
22 (U.S. EPA, 2008). Such areas tend to be dominated by relatively nonreactive bedrock in which
23 base cation production via weathering is limited (Elwood et al., 1991). The soils also usually
24 have thick organic horizons, high organic matter content in the mineral horizons, and low pH
25 (Joslin et al., 1992). Because of the largely nonreactive bedrock, base-poor litter and organic acid
26 anions produced by the conifers, high precipitation, and high leaching rates, soil base saturation
27 in these high elevation forests tends to be below 10%, and the soil cation exchange complex is
28 generally dominated by Al (Eagar et al., 1996; Johnson and Fernandez, 1992). The areas where
29 sugar maples appear to be at greatest risk are along ridges and where this species occurs on
30 nutrient-poor soils (U.S. EPA, 2008, Section 3.2.4). In addition, these forests support the growth
31 of sugar maple and red spruce, two species that are particularly sensitive to acidification.

1 Several characteristics were used to identify areas potentially sensitive to terrestrial
2 acidification. These characteristics included the following:

- 3 ▪ Soil depth
- 4 ▪ Bedrock composition
- 5 ▪ Soil pH
- 6 ▪ Presence of sugar maple or red spruce.

7 Geology is one of the most important factors in determining the potential sensitivity of an
8 area to terrestrial acidification (U.S. EPA, 2008, Section 3.2.4). In particular, the characteristics
9 of the soils and the upper portion of the bedrock can impact the buffering capacity of the soils in
10 a particular area. Acid-sensitive soils are those which contain low levels of exchangeable base
11 cations and low base saturation (U.S. EPA, 2008, Section 3.2.4).

12 It is important that soils be of sufficient depth for the rooting zone. Fine roots, which are
13 responsible for the vast majority of nutrient uptake, are typically concentrated in the upper 10 to
14 20 centimeters (cm) of soil (van der Salm and de Vries, 2001). These roots are most susceptible
15 to the impacts of acidification.

16 Bedrock composition and soil pH are two characteristics that are directly related to the
17 buffering capacity of a system. Soils overlying bedrock, such as calcium carbonate (e.g.,
18 limestone), which is reactive with acid, are more likely to successfully buffer acidifying
19 deposition than soils overlying nonreactive bedrock. In addition, soils with higher pH (i.e., more
20 alkaline) have a greater capacity to buffer acidifying deposition.

21 Areas with acid-sensitive geology were cross-referenced with the geographical ranges of
22 the ecological endpoints for this case study. As a result, locations with sugar maple or red spruce,
23 soil pH less than or equal to 5.0, soils less than or equal to 51 cm in depth, and low buffering
24 capacity bedrock (not dominated by carbonate rocks) were selected to represent areas with
25 potential sensitivity to acidification. A geographic information systems (GIS) analysis was
26 performed on datasets and data layers of physical, chemical, and biological properties to map
27 areas of potential sensitivity to acidification in the United States (**Figure 4.3-3**).

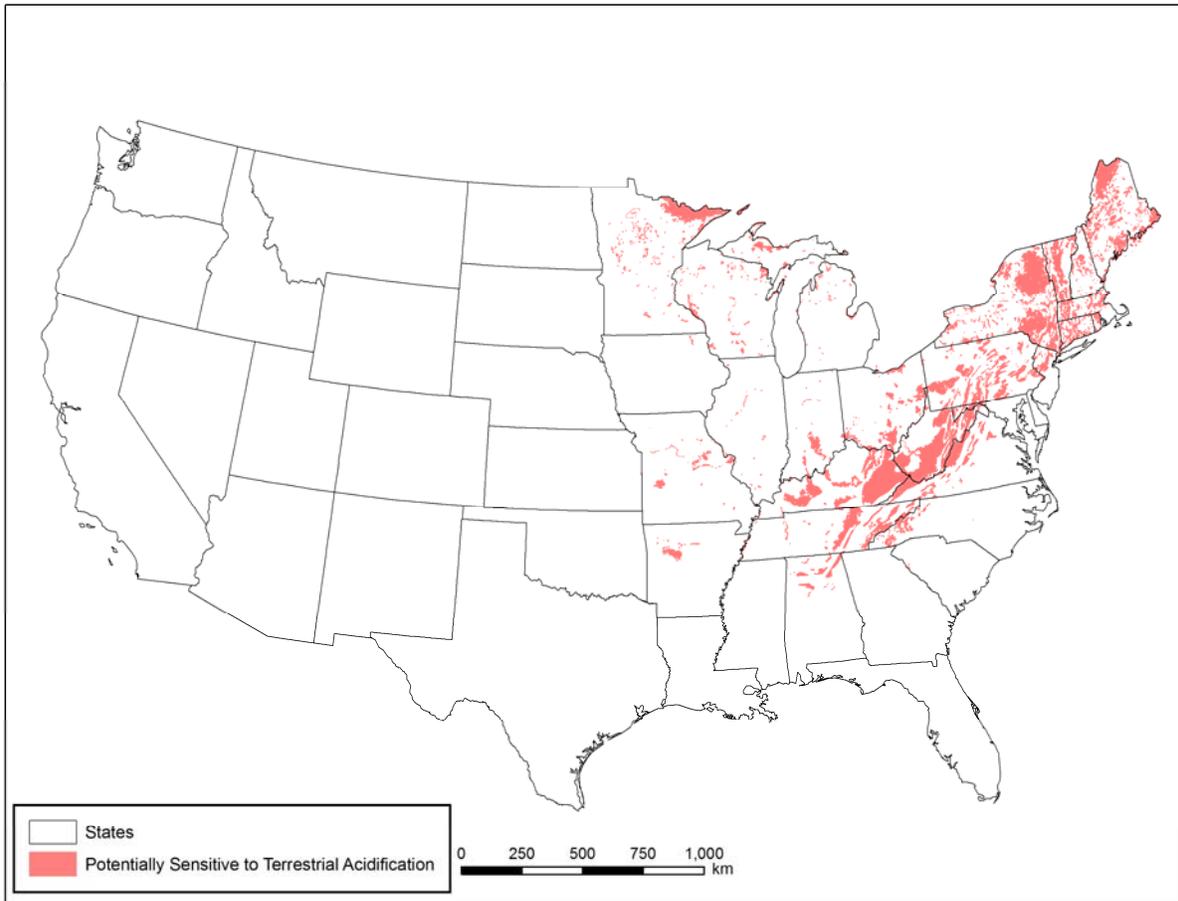
28 **4.3.3 Case Study Selection**

29 Following the identification of regions of potential sensitivity to acidification, risk and
30 exposure assessment sites recommended in the ISA (U.S. EPA, 2008, Appendix A) by the

1 Science Advisory Board – Ecological Effects Sub-committee (SAB-EES) (U.S. EPA, 2005) and
2 the body of published and unpublished literature were reviewed to determine the most suitable
3 areas for the red spruce and sugar maple case study areas.

4 Selection of an area for the sugar maple case study focused on the Allegheny Plateau
5 region in Pennsylvania, where a large proportion of published and unpublished research has been
6 focused. A significant amount of the research work in the Plateau region has been sponsored by
7 the United States Forest Service (USFS) and has produced extensive datasets of soil and tree
8 characteristics (Bailey et al., 2004; Hallett et al., 2006; Horsley et al., 2000). The USFS-
9 designated Kane Experimental Forest (KEF) was selected as the area for the sugar maple
10 terrestrial acidification case study. The KEF has been the focus of several long-term studies since
11 the 1930s. Seven plots (plot 1-plot 7) in the forest were assessed for this case study of the effects
12 of terrestrial acidification on sugar maples.

13 Selection of a case study area for red spruce involved the consideration of a variety of
14 regions. Four studies that examined the relationship between the Ca^{2+}/Al soil solution ratio and
15 tree health were identified, and relevant soil and tree information for each of the study regions
16 was compiled. A review of this information led to the selection of the Hubbard Brook
17 Experimental Forest (HBEF) in New Hampshire's White Mountains as the area for the red
18 spruce terrestrial acidification case study. The HBEF was also recommended in the ISA (U.S.
19 EPA, 2008, Appendix A) as a good area for risk and exposure assessment. This forest has
20 experienced high total nitrogen and sulfur deposition levels and low Ca^{2+}/Al soil solution ratios,
21 and has been the subject of extensive nutrient investigations and provided a large data set from
22 which to work on the case study. The case study of the effects of terrestrial acidification on red
23 spruce focused on Watershed 6 in the HBEF.



1
2 **Figure 4.3-3.** Map of areas of potential sensitivity of red spruce and sugar maple
3 to acidification in the United States (see Table 1.2-1 of Appendix 5 for a listing of
4 data sources to produce this map).

5 **4.3.4 Current Conditions Assessment**

6 The Simple Mass Balance (SMB) model, outlined in the International Cooperative
7 Programme (ICP) Mapping and Modeling Manual² (UNECE, 2004), was used to evaluate critical
8 loads of acidifying nitrogen and sulfur deposition in the KEF and HBEF case study areas,
9 according to Equation 7

$$10 \quad CL(S + N) = BC_{dep} - Cl_{dep} + BC_w - Bc_u + N_i + N_u + N_{de} - ANC_{le,crit} \quad (7)$$

11 where

12 $CL(S+N)$ = forest soil critical load for combined nitrogen and sulfur acidifying
13 deposition ($N+S_{comb}$)

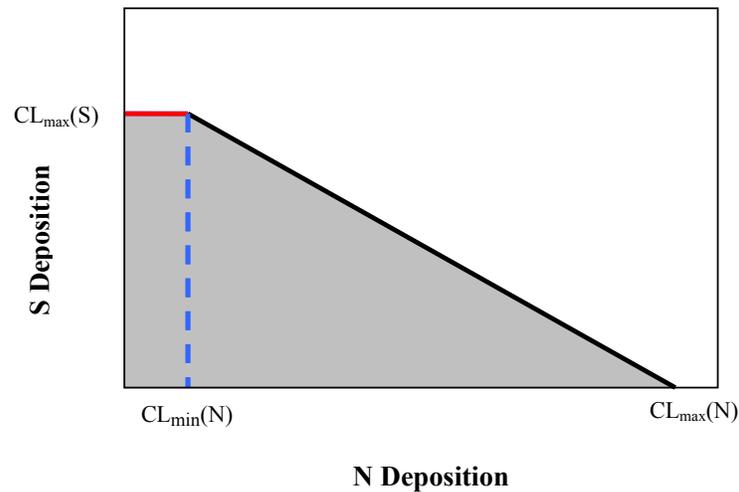
² The ICP Mapping and Modeling Manual (UNECE, 2004) recommends that wet deposition be corrected for sea salt on sites within 70 km of the coast. Neither the HBEF nor KEF case study areas are located less than 70 km from the coast, so this correction was not used.

- 1 BC_{dep} = base cation ($Ca^{2+} + K^+ + Mg^{2+} + Na^+$) deposition
 2 Cl_{dep} = chloride deposition
 3 BC_w = base cation ($Ca^{2+} + K^+ + Mg^{2+} + Na^+$) weathering
 4 Bc_u = uptake of base cations ($Ca^{2+} + K^+ + Mg^{2+}$) by trees
 5 N_i = nitrogen immobilization
 6 N_u = uptake of nitrogen by trees
 7 N_{de} = denitrification
 8 $ANC_{le,crit}$ = forest soil acid neutralizing capacity of critical load leaching

9 This model is currently one of the most commonly used approaches to estimate critical
 10 loads and has been widely applied in Europe (Sverdrup and de Vries, 1994), the United States
 11 (McNulty et al., 2007; Pardo and Duarte, 2007), and Canada (Arp et al., 2001; Ouimet et al.,
 12 2006; Watmough et al., 2006). It examines a long-term, steady-state balance of base cation,
 13 chloride, and nutrient inputs, “sinks,” and outputs within an ecosystem, and base cation
 14 equilibrium is assumed to equal the system’s critical load for ecological effects. A limitation of
 15 the SMB model is that it is a steady-state model and does not capture the cumulative changes in
 16 ecosystem conditions. However, as stated by the UNECE (2004), “Since critical loads are
 17 steady-state quantities, the use of dynamic models for the sole purpose of deriving critical loads
 18 is somewhat inadequate.” In addition, if a dynamic model is “used to simulate the transition to a
 19 steady state for the comparison with critical loads, care has to be taken that the steady-state
 20 version of the dynamic model is compatible with the critical load model.” Therefore, the
 21 selection of the SMB model was seen as the most suitable approach for this case study
 22 examining critical loads for sugar maple and red spruce.

23 A component of critical load determinations is the establishment of the critical load
 24 function (CLF). The CLF expresses the relationship between the critical load and all
 25 combinations of total nitrogen and sulfur deposition ($N+S_{comb}$) of an ecosystem. To define the
 26 CLF, minimum and maximum amounts of total nitrogen and sulfur deposition that combine to
 27 create the critical load must be determined (UNECE, 2004). The maximum amount of sulfur in
 28 the critical load ($CL_{max}(S)$) occurs when total nitrogen deposition does not exceed the nitrogen
 29 sinks (i.e., nitrogen immobilization, nitrogen uptake and removal by tree harvest, and
 30 denitrification) within the ecosystem. These nitrogen sinks are accounted for by the minimum
 31 amount of nitrogen in the critical load ($CL_{min}(N)$). Above this $CL_{min}(N)$ level, total nitrogen
 32 deposition can no longer be absorbed by the system, and acidification effects can occur. The
 33 maximum amount of nitrogen in the critical load ($CL_{max}(N)$) occurs when there is no sulfur
 34 deposition, and all of the acidity is due to the deposition of nitrogen.

1 An example of a CLF is depicted in **Figure 4.3-4**. All combinations of total nitrogen and
 2 sulfur deposition that fall on the black line representing the CLF are at the critical load. Any
 3 deposition combination that falls below the line or within the grey area is below the critical load.
 4 All combinations of nitrogen and sulfur deposition that are located above the line or within the
 5 white area are greater than the critical load.



6
 7 **Figure 4.3-4.** The critical load function created from the calculated maximum and
 8 minimum levels of total nitrogen and sulfur deposition (eq/ha/yr). The grey areas show
 9 deposition levels less than the established critical loads. The red line is the maximum
 10 amount of total sulfur deposition (valid only when nitrogen deposition is less than the
 11 minimum critical level of nitrogen deposition [blue dotted line]) in the critical load. The
 12 flat line portion of the curves indicates nitrogen deposition corresponding to the
 13 $CL_{\min}(N)$ (nitrogen absorbed by nitrogen sinks within the system).

14 **4.3.4.1 Input Data.**

15 This section summarizes the input data used in the calculations, the results for each case
 16 study area, and a comparison of these results with 2002 wet and dry nitrogen and sulfur
 17 deposition (combination of Community Multiscale Air Quality [CMAQ]-modeled 2002
 18 deposition results and 2002 National Atmospheric Deposition Program [NADP] deposition data).
 19 Additional detail, including an examination of the influence of different parameter values and
 20 methods, on the assessment of current conditions in the KEF and HBEF case study areas can be
 21 found in Appendix 5. Only the parameter values that were chosen to represent the current
 22 condition of the KEF and HBEF case study areas are presented here.

23 The majority of the data used to calculate critical loads for sugar maple and red spruce in
 24 the KEF and HBEF case study areas was specific to the case study areas and was compiled from
 25 published research studies and models, site-specific databases, or spatially-explicit GIS data

1 layers. However, several of the parameters (e.g., denitrification, nitrogen immobilization, the
2 gibbsite equilibrium constant, rooting zone soil depth) required the use of default values or
3 values used in published critical load assessments. Denitrification loss of nitrogen was assumed
4 to be 0 eq/ha/yr because both the KEF and HBEF study plots are upland forests, and
5 denitrification is considered negligible in such forests (McNulty et al. 2007; Ouimet et al., 2006;
6 Watmough et al., 2006). The nitrogen mobilization value was set to 42.86 eq/ha/yr for both
7 forests in this case study (McNulty et al., 2007). A $300 \text{ m}^6/\text{eq}^2$ value for the gibbsite equilibrium
8 constant (K_{gibb}) (used in the calculation of ANC) was selected because it is the most commonly
9 used default value (UNECE, 2004). Fifty centimeters (0.5 m) was selected as the rooting zone
10 soil depth for the forest soils of the two case study areas (Sverdrup and de Vries, 1994; Hodson
11 and Langan 1999). Base cation weathering (BC_w) rates were calculated using the clay-substrate
12 method (McNulty et al., 2007; Watmough et al., 2006). This is one of the most commonly used
13 methods to estimate base cation weathering for critical load analyses in North America. Base
14 cation (Bc_u) and nitrogen (N_u) uptake values were calculated in two different ways for the two
15 case study areas. In HBEF, Watershed 6 (the portion of HBEF evaluated for this case study) is a
16 reference watershed and does not have a history or future of harvesting; therefore, Bc_u and N_u
17 were assumed to be 0 eq/ha/yr. In KEF, the case study plots were assumed to be managed and
18 harvested on a regular basis. Values of Bc_u and N_u for this scenario were therefore calculated
19 using species-specific tree data and uptake estimates and were >0 eq/ha/yr. Three values of the
20 indicator of critical load, $(\text{Bc}/\text{Al})_{\text{crit}}$ soil solution ratio, were selected to represent different levels
21 of tree protection associated with total nitrogen and sulfur deposition: 0.6, 1.2, and 10 (**Table**
22 **4.3.2**). The $(\text{Bc}/\text{Al})_{\text{crit}}$ ratio of 0.6 represents the highest level of impact (lowest level of
23 protection) to tree health and growth and was selected because 75% of species found growing in
24 North America experience reduced growth at this Bc/Al ratio (**Figure 4.3.1**). In addition, a soil
25 solution Bc/Al ratio of 0.6 has been linked to a 20% and 35% reduction in sugar maple and red
26 spruce growth, respectively. The $(\text{Bc}/\text{Al})_{\text{crit}}$ ratio of 1.2 is considered to represent a moderate
27 level of impact, as the growth of 50% of tree species (found growing in North America) was
28 negatively impacted at this soil solution ratio. The $(\text{Bc}/\text{Al})_{\text{crit}}$ ratio of 10.0 represents the lowest
29 level of impact (greatest level of protection) to tree growth; it is the most conservative value used
30 in studies that have calculated critical loads in the United States and Canada (Canada (McNulty
31 et al. 2007; NEG/ECP, 2001; Watmough et al., 2004).

Table 4.3-2. The Three Indicator $(Bc/Al)_{crit}$ Soil Solution Ratios and Corresponding Levels of Protection to Tree Health and Critical Loads

Indicator $(Bc/Al)_{crit}$ Soil Solution Ratio	Level of Protection to Tree Health	Critical Load
0.6	Low	High
1.2	Intermediate	Intermediate
10.0	High	Low

1

2 **4.3.5 Results for the Case Study Areas**

3 Based on the input data described above, the three critical loads for the KEF case study
4 area, in order of lowest to highest protection level, were 2,009, 1,481 and 910 eq/ha/yr (for
5 $Bc/Al_{(crit)} = 0.6, 1.2,$ and 10.0, respectively). In the HBEF Case Study Area, these values, in the
6 same order of protection, were 1,237, 892, and 487 eq/ha/yr (for $Bc/Al_{(crit)} = 0.6, 1.2,$ and 10.0,
7 respectively).

8 The $(Bc/Al)_{crit}$ ratio of 0.6 represents the highest level of impact (lowest level of
9 protection) to tree health and growth; as much as 75% of 46 tree species found in North America
10 experience reduced growth at this ratio (Sverdrup and Warfvinge, 1993). Both red spruce and
11 sugar maple show at least a 20% reduction in growth at the 0.6 $(Bc/Al)_{crit}$ ratio.

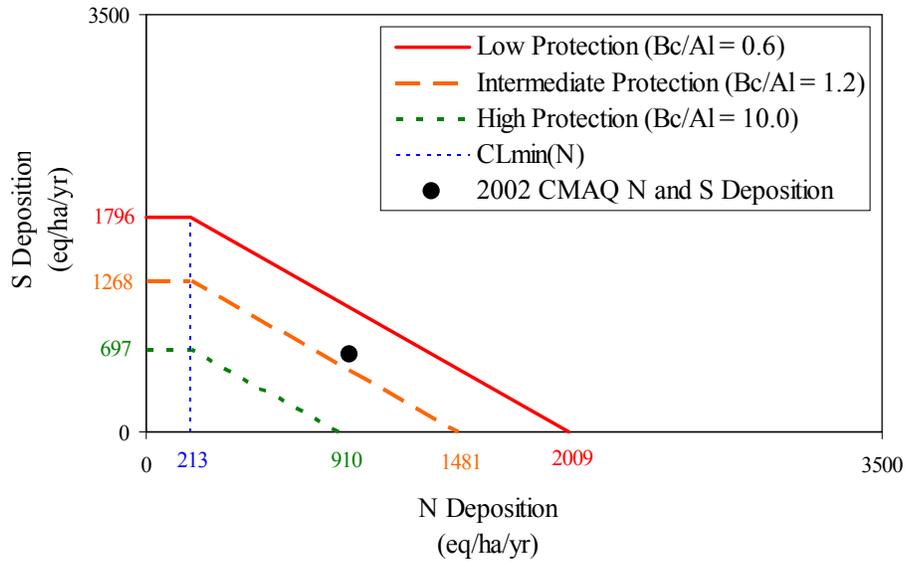
12 ***Comparison with 2002 Deposition Data***

13 This section discusses the impact of 2002 CMAQ/NADP total nitrogen and sulfur
14 deposition relative to the estimated critical loads at the KEF and HBEF case study areas.
15 According to 2002 CMAQ/NADP total nitrogen and sulfur deposition, the KEF Case Study Area
16 received 13.6 kg N/ha (967.5 eq/ha) and 20.7 kg S/ha (646.4 eq/ ha), and the HBEF Case Study
17 Area experienced 8.4 kg N/ha (601.1 eq/ha) and 7.5 kg S/ha (233.1 eq/ha).

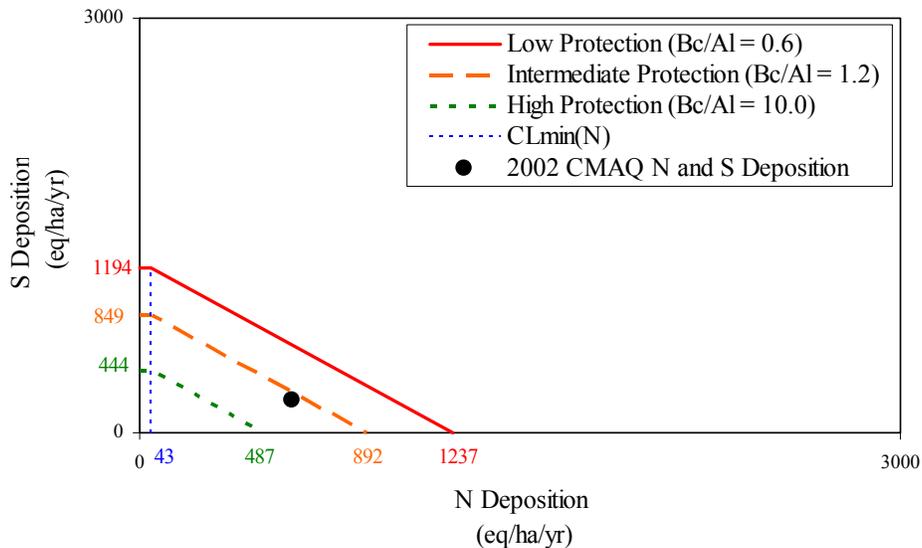
18 As outlined above, 2,009, 1,481, and 910 eq/ha/yr were the critical loads selected to
19 represent the three levels of protection for the KEF Case Study Area and 1,237, 892, and 487
20 eq/ha/yr were the critical loads selected for the HBEF Case Study Area. These estimates are
21 based on the critical load parameters suggested and most frequently used by scientists and
22 previous research. When compared to the 2002 CMAQ/NADP total nitrogen and sulfur
23 deposition levels, it was evident that the deposition levels were greater than the most protective
24 critical load ($Bc/Al_{(crit)} = 10.0$) for both case study areas and also greater than the intermediate
25 protection critical load ($Bc/Al_{(crit)} = 1.2$) for the KEF Case Study Area (**Figure 4.3-5** and **Figure**

1 **4.3-6).** In these comparisons, total nitrogen and sulfur deposition exceeded the KEF Case Study
2 Area critical load by 132 – 704 eq/ha/yr and exceeded the HBEF Case Study Area’s critical load
3 by 347 eq/ha/yr. Similar results have been reported in other studies that have assessed the two
4 case study areas. McNulty et al. (2007) and Pardo and Driscoll (1996) found that deposition
5 levels were greater than the estimated critical loads in the HBEF Case Study Area. McNulty et
6 al. (2007) also reported that total nitrogen and sulfur deposition in the KEF exceeded the
7 calculated critical loads for the case study area in KEF. These results suggest that the health of
8 red spruce at HBEF and sugar maple at KEF may have been compromised by the acidifying
9 nitrogen and sulfur deposition received in 2002.

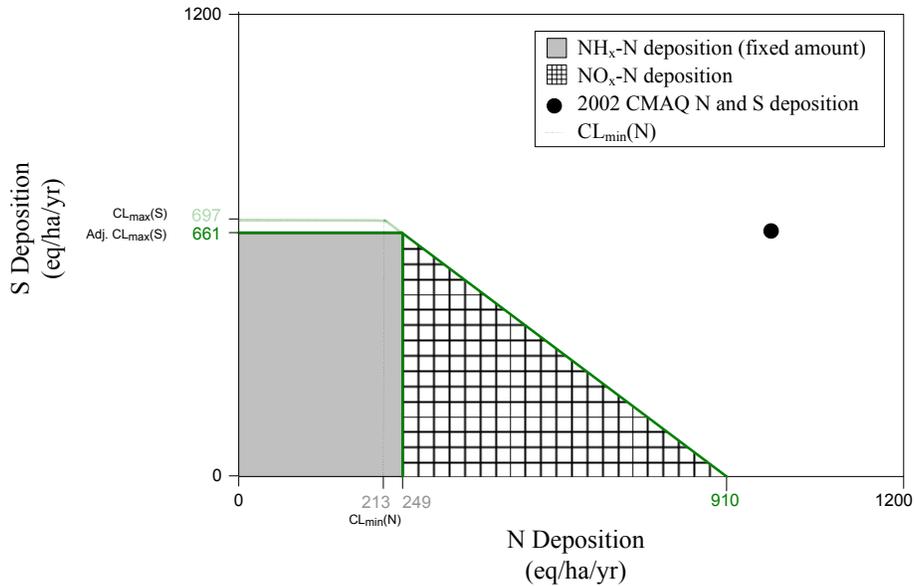
10 Acidifying total nitrogen deposition consists of both reduced (NH_x) and oxidized (NO_x)
11 forms of nitrogen. However, only NO_x is currently regulated as a criteria pollutant. Therefore, to
12 gain an understanding of the relationship between the two states (reduced and oxidized) of total
13 nitrogen deposition and the critical loads for the KEF and HBEF case study areas, total nitrogen
14 deposition must be separated into NH_x -N and NO_x -N. **Figure 4.3-7** and **Figure 4.3-8** present the
15 CLF response curves for the most protective critical load condition ($\text{Bc}/\text{Al}_{(\text{crit})} = 10.0$) for the
16 KEF and HBEF case study areas, respectively. In these relationships, the CLF function has been
17 modified by maintaining NH_x -N deposition at the 2002 deposition level; only sulfur and NO_x -N
18 deposition levels vary to indicate the combined critical load. Based on 2002 CMAQ/NADP total
19 nitrogen and sulfur deposition, NH_x -N accounted for 25.7 % (249 eq/ha) and 26.4 % (159 eq/ha)
20 of total nitrogen deposition in the KEF and HBEF case study areas, respectively. These fixed
21 amounts of NH_x -N influenced the highest protection CLF response curves for both areas. For
22 both case studies, the maximum sulfur critical load ($\text{CL}_{\text{max}}(\text{S})$) and the maximum nitrogen
23 critical load ($\text{CL}_{\text{max}}(\text{N})$), as NO_x , were lowered. In the calculations for the KEF Case Study Area,
24 the $\text{CL}_{\text{max}}(\text{S})$ was reduced by 5 % to 661 eq/ha/yr, and in the HBEF Case Study Area
25 calculations, the $\text{CL}_{\text{max}}(\text{S})$ was reduced by 26 % to 328 eq/ha/yr. Similarly, the $\text{CL}_{\text{max}}(\text{N})$ (as
26 NO_x) for the KEF Case Study Area was reduced by 27% to 661 eq/ha/yr, and the $\text{CL}_{\text{max}}(\text{N})$ (as
27 NO_x) for the HBEF Case Study Area was reduced by 33% to 328 eq/ha/yr when NH_x -N
28 deposition was held constant.



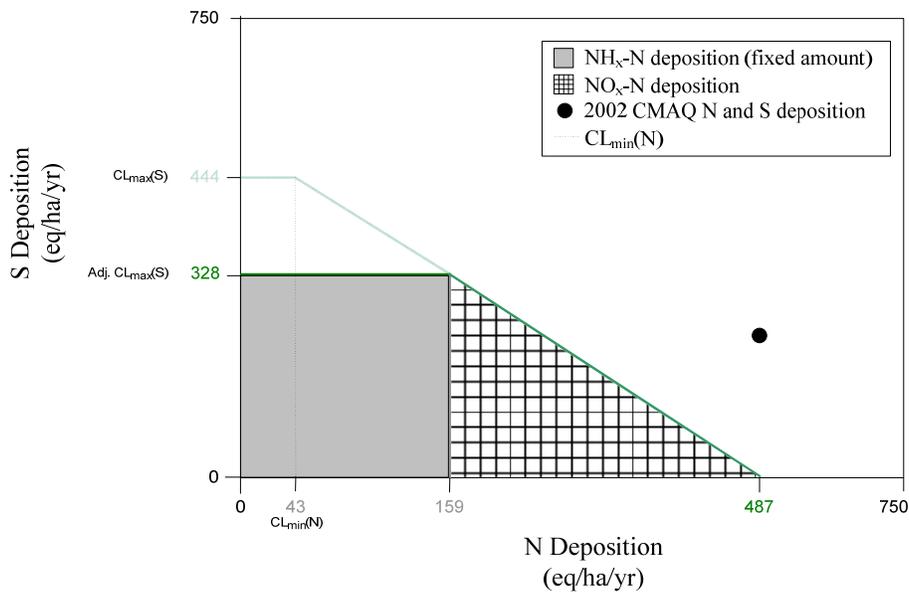
1
2 **Figure 4.3-5.** Critical load function response curves for the three selected critical
3 loads conditions (corresponding to the three levels of protection) for the Kane
4 Experimental Forest Case Study Area. The 2002 CMAQ/NADP total nitrogen and
5 sulfur ($N+S_{comb}$) deposition was greater than the highest and intermediate level of
6 protection critical loads. The flat line portion of the curves indicates total nitrogen
7 deposition corresponding to the $CL_{min}(N)$ (nitrogen absorbed by nitrogen sinks within
8 the system).



9
10 **Figure 4.3-6.** Critical load function response curves for the three selected critical
11 loads conditions (corresponding to the three levels of protection) for the Hubbard
12 Brook Experimental Forest Case Study Area. The 2002 CMAQ/NADP total
13 nitrogen and sulfur ($N+S_{comb}$) deposition was greater than the highest level of
14 protection critical load. The flat line portion of the curves indicates total nitrogen
15 deposition corresponding to the $CL_{min}(N)$ (nitrogen absorbed by nitrogen sinks
16 within the system).



1
 2 **Figure 4.3-7.** The influence of the 2002 CMAQ/NADP total reduced nitrogen
 3 ($\text{NH}_x\text{-N}$) deposition on the critical function response curve, and in turn, the
 4 maximum amounts of sulfur ($\text{CL}_{\text{max}}(\text{S})$) and oxidized nitrogen ($\text{NO}_x\text{-N}$) in the
 5 critical load for the Kane Experimental Forest Case Study Area. The critical load
 6 of oxidized nitrogen ($\text{NO}_x\text{-N}$) is 661 eq/ha/yr (910–249). The $\text{CL}_{\text{min}}(\text{N})$ (nitrogen
 7 absorbed by nitrogen sinks within the system) corresponds to the value depicted
 8 in **Figure 4.3-5**.



9
 10 **Figure 4.3-8.** The influence of the 2002 CMAQ/NADP total reduced nitrogen
 11 ($\text{NH}_x\text{-N}$) deposition on the critical load function response curve and, in turn, the
 12 maximum amounts of sulfur ($\text{CL}_{\text{max}}(\text{S})$) and oxidized nitrogen ($\text{NO}_x\text{-N}$) in the
 13 critical load for the Hubbard Brook Experimental Forest Case Study Area. The
 14 critical load of oxidized nitrogen ($\text{NO}_x\text{-N}$) is 328 eq/ha/yr (487–159). The
 15 $\text{CL}_{\text{min}}(\text{N})$ (nitrogen absorbed by nitrogen sinks within the system) corresponds to
 16 the value depicted in **Figure 4.3-6**.

4.3.6 Evaluation of Representativeness of Case Study Areas

Although the case studies estimated critical load assessments for red spruce and sugar maple in two areas and established that 2002 CMAQ/NADP total nitrogen and sulfur deposition was greater than the calculated loads, these results cannot be directly extrapolated to the full ranges of the two species. Critical loads are largely determined by soil characteristics, and these characteristics vary by location. Therefore, to characterize the critical loads of sugar maple and red spruce and determine the extent to which total nitrogen and sulfur deposition is greater than or less than these loads, it is necessary to calculate critical loads in multiple locations throughout the ranges of the two species to determine the critical loads for these species.

Critical load calculations were applied to multiple areas within 24 states for sugar maple and in 8 states for red spruce. Individual site locations within each state were determined by the U.S. Forest Service Forest Inventory and Analysis (FIA) database permanent sampling plots' locations on forestland³ (timberland⁴ for New York, Arkansas, Kentucky, and North Carolina), each covering 0.07 ha. Only database information for nonunique⁵, permanent sampling plots that supported the growth of sugar maple or red spruce and had the necessary soil, parent material, atmospheric deposition, and runoff data were included in the analyses. With these restrictions, 7,992 of the 14,669 sugar maple plots and 763 of the 2,875 red spruce plots were included in the calculations of the plot-specific critical loads (**Table 4.3-3**). Although only a subset of the total sugar maple and red spruce plots were included in the analyses, the results are thought to accurately capture the range and trends of critical loads of the two species. Because of the randomness of the plot restrictions, it is unlikely that a bias was incorporated into the analyses.

The calculated critical loads for the three levels of protection ($Bc/AI_{(crit)} = 0.6, 1.2, \text{ and } 10.0$) for all plots were compared to 2002 CMAQ/NADP total nitrogen and sulfur deposition to

³ Forestland is defined as, "land at least 10 percent stocked by forest trees of any size, or formerly having such tree cover, and not currently developed for non-forest uses, with a minimum area classification of 1 acre." (USFS, 2002a).

⁴ Timberland is defined as, "forest land capable of producing in excess of 20 cubic feet per acre per year and not legally withdrawn from timber production, with a minimum area classification of 1 acre." (USFS, 2002b).

⁵ Nonunique permanent sampling plot locations are those that have critical load attribute values (e.g., soils, runoff, atmospheric deposition) that are not distinct and are repeated within a 250-acre area of the plot location. This "confidentiality" filter is a requirement of the USFS to prevent the disclosure of data that can be directly linked to a location on private land. To comply with the necessary "confidentiality," full coverages of the data required for the critical load deposition calculations were given to the USFS, and the USFS matched and provided the data to each nonunique, permanent sampling plot.

- 1 determine which plots with sugar maple and/or red spruce experienced deposition levels greater
- 2 than the critical load values.

Table 4.3-3. Number and Location of USFS FIA Permanent Sampling Plots Used in the Analysis of Critical Loads for Full Ranges of Sugar Maple and Red Spruce

State	Sugar Maple	Red Spruce
Alabama	13	–
Arkansas	10	–
Connecticut	35	–
Illinois	29	–
Indiana	306	–
Iowa	13	–
Kansas	NA	–
Kentucky	14	–
Maine	271	560
Maryland	4	–
Massachusetts	33	3
Michigan	633	–
Minnesota	289	–
Missouri	147	–
New Hampshire	82	55
New Jersey	6	–
New York	485	52
North Carolina	17	1
Ohio	374	–
Pennsylvania	285	NA
Rhode Island	NA	–
South Carolina	NA	–
Tennessee	319	1
Vermont	114	11
Virginia	175	NA
West Virginia	378	7
Wisconsin	960	–
TOTAL	4,992	763

NA = data not available for state

“–” = tree species not present on forestland in state

3
4

4.3.7 Current Conditions for Sugar Maple and Red Spruce

The critical loads of acidifying deposition for sugar maple in 24 states for the three levels of protection were found to range from 107 to 6,008 eq/ha/yr (**Table 4.3-4**). Critical loads for red spruce in the 8 states ranged from 180 to 4,278 eq/ha/yr. In a comparison of the 2002 CMAQ/NADP total nitrogen and sulfur deposition levels and calculated critical loads, 3% to 75% of all sugar maple plots and 3% to 36% of all red spruce plots were found to have total nitrogen and sulfur deposition greater than the critical loads; the highest protection critical loads ($Bc/Al_{(crit)} = 10.0$) had the highest frequency of exceedance (**Table 4.3-5**). Aggregated by state, a large proportion of the sugar maple and red spruce plots showed high levels of critical load exceedance for the highest protection level ($Bc/Al_{(crit)} = 10.0$), and comparatively lower exceedance frequency at the lowest protection level ($Bc/Al_{(crit)} = 0.6$) (**Table 4.3-5**). In general, New Hampshire displayed the greatest degree of critical load exceedance at all protection levels for both species.

Collectively, these results suggest that the health of at least a portion of the sugar maple and red spruce growing in the United States may have been compromised with the acidifying total nitrogen and sulfur deposition in 2002; even with the lowest level of protection, half the states contained sugar maple and red spruce stands that were negatively impacted by acidifying deposition. At the highest level of protection ($Bc/Al_{(crit)} = 10.0$), the apparent impact of the 2002 CMAQ/NADP total nitrogen and sulfur deposition was much greater. A large portion of sugar maple (i.e., $\geq 80\%$ of plots in 13 of 24 states) and the majority of red spruce (i.e., 100% of plots in 5 of 8 states) experienced deposition levels that exceeded the critical loads. If this high protection critical load accurately represents the conditions of the two species, a large proportion of both sugar maple and red spruce, throughout their ranges, were most likely negatively impacted by total nitrogen and sulfur deposition in 2002.

Table 4.3-4. Ranges of Critical Load Values, by Level of Protection ($Bc/Al_{(crit)} = 0.6, 1.2,$ and 10.0) and by State, for the Full Distribution Ranges of Sugar Maple and Red Spruce

State	Ranges of Critical Load Values (eq/ha/yr)					
	Sugar Maple			Red Spruce		
	$Bc/Al = 0.6$	$Bc/Al = 1.2$	$Bc/Al = 10.0$	$Bc/Al = 0.6$	$Bc/Al = 1.2$	$Bc/Al = 10.0$
Alabama	1,592 to 5,337	1,114 to 3,638	617 to 2,015	–	–	–

State	Ranges of Critical Load Values (eq/ha/yr)					
	Sugar Maple			Red Spruce		
	Bc/Al = 0.6	Bc/Al = 1.2	Bc/Al = 10.0	Bc/Al = 0.6	Bc/Al = 1.2	Bc/Al = 10.0
Arkansas	2,239 to 4,290	1,536 to 2,913	857 to 1,623	–	–	–
Connecticut	1,519 to 2,468	1,058 to 1,702	581 to 941	–	–	–
Illinois	2,543 to 3,671	1,730 to 2,485	965 to 1,390	–	–	–
Indiana	1,478 to 5,859	1,020 to 3,971	573 to 2,214	–	–	–
Iowa	2,260 to 3,791	1,533 to 2,560	854 to 1,424	–	–	–
Kansas	NA	NA	NA	–	–	–
Kentucky	2,044 to 3,994	1,390 to 2,707	749 to 1,497	–	–	–
Maine	746 to 4,284	535 to 2,983	295 to 1,620	599 to 4,278	439 to 2,979	249 to 1,623
Maryland	2,066 to 3,090	1,417 to 2,122	929 to 1,178	–	–	–
Massachusetts	791 to 2,414	566 to 1,661	319 to 919	1,706 to 1,736	1,191 to 1,213	656 to 669
Michigan	400 to 6,008	294 to 4,070	169 to 2,269	–	–	–
Minnesota	220 to 4,916	166 to 3,318	107 to 1,861	–	–	–
Missouri	978 to 4,891	681 to 3,304	377 to 1,843	–	–	–
New Hampshire	580 to 1,994	419 to 1,439	236 to 780	418 to 1,994	324 to 1,439	180 to 780
New Jersey	1,452 to 2,651	1,029 to 1,824	566 to 1,012	–	–	–
New York	503 to 4,467	370 to 3,039	209 to 1,686	526 to 3,146	386 to 2,156	217 to 1,195
North Carolina	1,415 to 3,444	1,010 to 2,426	558 to 1,319	1256	926	501
Ohio	1,226 to 4,986	855 to 3,366	469 to 1,877	–	–	–
Pennsylvania	1,026 to 4,047	723 to 2,752	402 to 1,530	NA	NA	NA
Rhode Island	NA	NA	NA	–	–	–
South Carolina	NA	NA	NA	–	–	–
Tennessee	921 to 5,755	653 to 3,901	351 to 2,175	2,065	1,433	788
Vermont	479 to 5,660	351 to 3,846	201 to 2,142	1,462 to 2,141	1,036 to 1,534	574 to 825
Virginia	1,036 to 5,852	726 to 3,968	410 to 2,208	NA	NA	NA
West Virginia	369 to 4,134	270 to 2,819	152 to 1,560	2,300 to 3,634	1,610 to 2,533	884 to 1,382
Wisconsin	400 to 5,031	290 to 3,393	166 to 1,898	–	–	–
Combined (all plots)	220 to 6,008	166 to 4,070	107 to 2,269	418 to 4,278	324 to 2,979	180 to 1,623

1 NA = data not available for state

2 “–” = tree species not present on forestland in state

3

Table 4.3-5. Percentages of Plots, by Protection Level ($Bc/Al_{(crit)} = 0.6, 1.2, \text{ and } 10.0$) and by State, Where 2002 CMAQ/NADP Total Nitrogen and Sulfur Deposition Was Greater Than the Critical Loads for Sugar Maple and Red Spruce

State	Percentage of Plots Where Critical Load is Exceeded (%)					
	Sugar Maple			Red Spruce		
	Bc/Al = 0.6	Bc/Al = 1.2	Bc/Al = 10.0	Bc/Al = 0.6	Bc/Al = 1.2	Bc/Al = 10.0
Alabama	0	23	31	–	–	–
Arkansas	0	0	10	–	–	–
Connecticut	0	23	100	–	–	–
Illinois	0	0	66	–	–	–
Indiana	0.3	12	87	–	–	–
Iowa	0	0	23	–	–	–
Kansas	NA	NA	NA	–	–	–
Kentucky	0	0	86	–	–	–
Maine	0	0.7	20	0.2	0.5	16
Maryland	0	25	100	–	–	–
Massachusetts	6	33	100	0	100	100
Michigan	6	14	70	–	–	–
Minnesota	2	7	30	–	–	–
Missouri	0.7	2	46	–	–	v
New Hampshire	29	38	84	27	38	78
New Jersey	0	67	100	–	–	–
New York	6	20	95	14	15	79
North Carolina	0	6	71	0	0	100
Ohio	1	16	95	–	–	–
Pennsylvania	7	22	98	NA	NA	NA
Rhode Island	NA	NA	NA	–	–	–
South Carolina	NA	NA	NA	–	–	–
Tennessee	0.3	3	50	0	0	100
Vermont	2	7	99	2	6	100
Virginia	2	9	59	NA	NA	NA
West Virginia	2	8	95	0	0	100
Wisconsin	2	10	82	–	–	–
Combined (all plots)	3	12	75	3	5	36

1 NA = data not available for state

2 “–” = tree species not present on forestland in state

4.3.8 Ecological Effect Function for Terrestrial Acidification

As described earlier and explained in greater detail in Appendix 5, there is an established relationship between atmospheric deposition of nitrogen and sulfur and the Bc/Al ratio in the soil solution. In areas with high amounts of acidifying nitrogen and sulfur deposition, protons can replace exchangeable base cations, which are then leached from the soil, and the resulting lower soil pH increases the mobilization of soil Al. The Bc/Al ratio in the soil solution is thereby decreased, and this can negatively impact trees through direct Al toxicity to roots and reduced uptake of base cation nutrients. As indicated in **Figure 4.3.1**, as the Bc/Al ratio in the soil solution decreases, the incidence of reduced tree growth increases.

The Bc/Al ratio in the soil solution was the indicator selected to estimate critical loads of acidity for terrestrial acidification and is an influential parameter in the ANC term of the SMB model critical load equation (Equation 7 in Section 4.3.4). The equation to estimate ANC is presented below, in Equation 8.

$$\text{ANC}_{(\text{le,crit})} = -Q^{2/3} \times \left(1.5 \times \frac{\text{Bc}_{\text{dep}} + \text{Bc}_{\text{w}} - \text{Bc}_{\text{u}}}{K_{\text{gibb}} \times \left(\frac{\text{Bc}}{\text{Al}} \right)_{\text{crit}}} \right)^{1/3} - 1.5 \times \frac{\text{Bc}_{\text{dep}} + \text{Bc}_{\text{w}} - \text{Bc}_{\text{u}}}{\left(\frac{\text{Bc}}{\text{Al}} \right)_{\text{crit}}} \quad (8)$$

where

Q	=	annual runoff in m ³ /ha/yr
Bc_{dep}	=	base cation (Ca ²⁺ + K ⁺ + Mg ²⁺) deposition ⁶
Bc_{w}	=	soil base cation (Ca ²⁺ + K + Mg ²⁺) weathering ⁷
Bc_{u}	=	base cation (Ca ²⁺ + K + Mg ²⁺) uptake by trees
K_{gibb}	=	the gibbsite equilibrium constant (a function of forest soil organic matter content that affects Al solubility) (UNECE, 2004)
$(\text{Bc}/\text{Al})_{\text{crit}}$	=	the base cation to aluminum ratio (indicator)

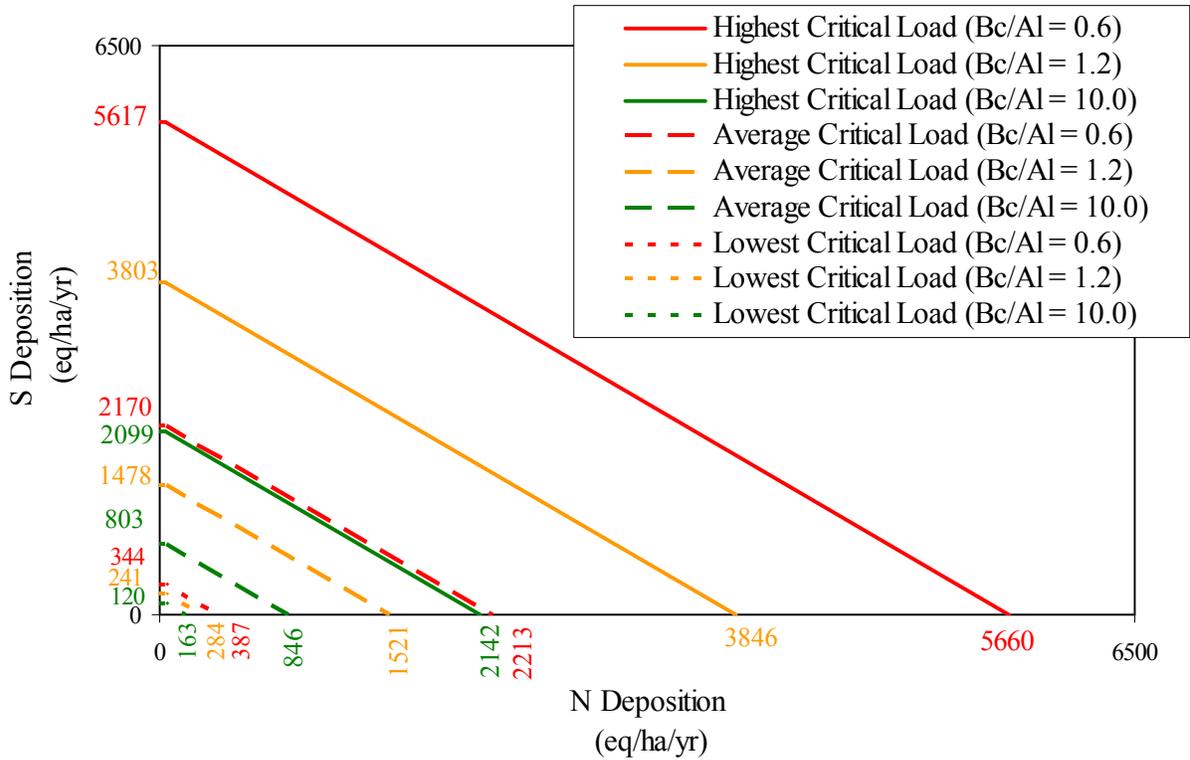
The three $(\text{Bc}/\text{Al})_{\text{crit}}$ ratios (0.6, 1.2, and 10.0) used in this case study were selected to represent the range of protection levels to the health of red spruce and sugar maple. The Bc/Al(crit) ratio of 10.0 corresponds to the highest level of protection, and, when included in the calculation of the ANC term, results in the lowest critical load. A terrestrial system with such a

⁶ Bc_{dep} is **not** the same as BC_{dep} used in Equation 1. BC_{dep} includes Ca²⁺, K⁺, Mg²⁺, and Na⁺, whereas Bc_{dep} includes base cations that are taken up by vegetation (i.e., only includes Ca²⁺, K⁺, and Mg²⁺).

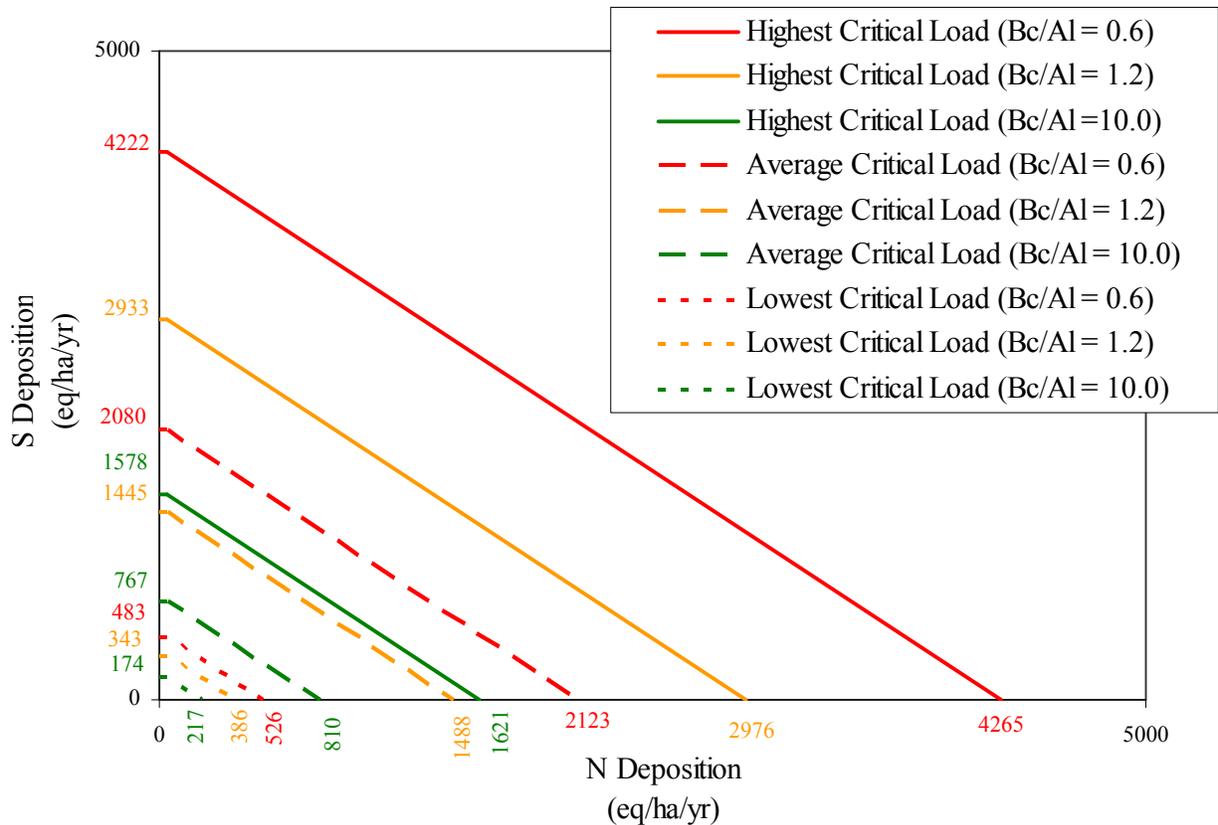
⁷ Bc_{w} is **not** the same as BC_{w} used in Equation 1. BC_{w} includes Ca²⁺, K⁺, Mg²⁺, and Na⁺, whereas Bc_{w} includes base cations that are taken up by vegetation (i.e., only includes Ca²⁺, K⁺, and Mg²⁺).

1 condition would only be able to tolerate comparatively low levels of total nitrogen and sulfur
2 deposition. A $(Bc/Al)_{crit}$ ratio of 1.2 represents an intermediate level of protection and moderate
3 critical load. The $(Bc/Al)_{crit}$ ratio of 0.6 ratio provides the lowest level of protection to tree
4 health and results in the estimation of a high critical load.

5 In the expansion of the critical load assessments to the full ranges of sugar maple and red
6 spruce, as discussed in Section 4.3.6, critical loads were estimated in multiple locations for each
7 of the three levels of protection ($(Bc/Al)_{crit} = 0.6, 1.2, \text{ and } 10.0$) for each species. Because of the
8 differences in soil conditions, runoff, base cation, and chloride deposition patterns, this analysis
9 produced a wide range of critical load estimates. Depicting the extremes (lowest and highest) and
10 the average critical load values in CLF curves provides an indication of the combinations of total
11 nitrogen and sulfur deposition that could occur without exceeding the critical loads associated
12 with the upper and lower limits and averages of the three protection levels (**Figure 4.3.9** and
13 **Figure 4.3.10**). As is depicted in the figures, the lowest critical loads corresponding to the three
14 protection levels ($(Bc/Al)_{crit}$ ratio = 0.6, 1.2, and 10.0) were 387, 284, and 163 eq/ha/yr for sugar
15 maple and 526, 386, and 217 eq/ha/yr for red spruce. In contrast, the highest critical loads for the
16 three protection levels for sugar maple were 5,660, 3,846, and 2,142 eq/ha/yr and for red spruce
17 were 4,265, 2,976, and 1621 eq/ha/yr. The average critical loads associated with the three levels
18 of protection were 2,213, 1,521, and 846 eq/ha/yr for sugar maple and 2,123, 1,488, and 810
19 eq/ha/yr for red spruce. The 1,979 to 5,273 eq/ha/yr differences between the extreme estimates
20 (i.e., highest and lowest critical loads) for sugar maple and 1,404 to 2,590 eq/ha/yr differences
21 for the red spruce estimates indicate the amount of total nitrogen and sulfur deposition that
22 separates the lower and upper limits of the lowest and highest protection levels.



1
 2 **Figure 4.3.9.** The extreme (i.e., highest and lowest) and average critical load
 3 function response curves for the three levels of protection ($(Bc/Al)_{crit} = 0.6, 1.2,$
 4 and 10.0) for the critical load assessments for the full range of sugar maple. The
 5 $CL_{min}(N)$ value for all curves is 42.86 eq/ha/yr, but it is not shown in the figure.



1
2 **Figure 4.3.10.** The extreme (i.e., highest and lowest) and average critical load
3 function response curves for the three levels of protection ($(Bc/Al)_{crit} = 0.6, 1.2,$
4 and 10.0) for the critical load assessments for the full range of red spruce. The
5 $CL_{min}(N)$ value for all curves is 42.86 eq/ha/yr, but it is not shown in the figure.

6 4.3.9 Uncertainty and Variability

7 4.3.9.1 Kane Experimental Forest and Hubbard Brook Experimental Forest Case 8 Study Areas

1 Despite the extensive use of the SMB model
2 to estimate critical loads, there is uncertainty
3 regarding the output from the model and calculations.
4 To a large degree, this uncertainty comes from the
5 dependence of the SMB calculations on assumptions
6 made by the researcher and the use of default values.
7 Parameters including base cation weathering, forest
8 soil ANC of critical load leaching, K_{gibb} , base cation
9 and nitrogen uptake, nitrogen immobilization, and
10 denitrification are rarely measured at each location
11 and must be selected based on the literature or on
12 other calculations and models. In an analysis
13 conducted by Li and McNulty (2007), it was
14 determined that the base cation weathering and forest soil ANC of critical load leaching
15 parameters were the main sources of uncertainty, with each respectively contributing 49% and
16 46% to the total variability in critical load estimates. It has, therefore, been suggested that the
17 calculation of critical loads using a relevant range of parameter values can provide the
18 foundation for an uncertainty analysis (Hall et al., 2001; Hodson and Langan 1999; Li and
19 McNulty, 2007;); it is likely that the correct critical load of a system will be contained within the
20 range of load estimates from such an approach. If all or a large majority of estimates indicate that
21 the critical load of a system is exceeded with current total nitrogen and sulfur deposition rates,
22 the probability is high that deposition is greater than the critical load and that the trees and
23 vegetation in that system are being negatively impacted by acidification. Conversely, if
24 deposition is not greater than the majority of critical load estimates, there can be greater
25 confidence that the system is not being impacted by acidifying deposition. Under a scenario of a
26 near equal number of estimates indicating exceedance and nonexceedance, however, there is low
27 probability that the actual acidification status of a system can be accurately determined.
28 Nonetheless, such results do suggest that the system is near the critical load level and should be
29 monitored or assessed more thoroughly.

“Uncertainty is a measure of the knowledge of the magnitude of a parameter. Uncertainty can be reduced by research, i.e., the parameter value can be refined. Uncertainty is quantified as a distribution. For example, the volume of a lake may be estimated from its surface area and an average depth. This estimate can be refined by measurement. Variance is a measure of the heterogeneity of a landscape parameter or the inherent variability in a chemical property. Variance can not be reduced by further research. It is quantified as a distribution. For example, the organic carbon content of the soil in a region may vary, even over short distances. The soil is not homogenous and thus the organic carbon content can be described with a distribution of values” (Webster and MacKay, 2003).

30 In this case study, multiple values were used for several parameters in the SMB
31 calculations and are detailed in Appendix 5. Therefore, it was possible to use the range of output
32 values from the calculations to assess the certainty of the acidification status of the HBEF and

1 KEF case study areas. The patterning of the results suggest that the 2002 total nitrogen and sulfur
2 deposition levels were very close to, if not greater than, the critical loads of the two case study
3 areas, and both ecosystems are likely to be sensitive to any future changes in the levels of
4 nitrogen and sulfur acidifying deposition.

5 ***4.3.9.2 Expansion of Critical Load Assessments to Determine Current Conditions for*** 6 ***Sugar Maple and Red Spruce***

7 Critical load estimates for individual plots within the distribution ranges of sugar maple
8 and red spruce were calculated using the clay-substrate method to estimate BC_w . As discussed
9 earlier, the BC_w term within the SMB model is one of the most influential terms in the
10 calculation of a critical load, and the determination of this BC_w value is strongly influenced by
11 the classified acidity of the soil parent material. In large-scale analyses, descriptions of the
12 mineralogy of parent material underlying the soil may be missing, nondescriptive, only
13 suggestive of mineralogy, or these may only represent the dominant mineralogy in a large area
14 (and therefore not accurately capture the smaller-scale variation in mineralogy). Therefore, it is
15 possible to misclassify the parent material acidity in the BC_w term.

16 In the analyses of critical loads for the full distribution ranges of sugar maple and red
17 spruce in this Risk and Exposure Assessment, two fine-scale databases, the Soil Survey
18 Geographic Database (SSURGO) of soils [USDA-NRCS, 2008] and USGS state-level geology
19 [USGS, 2009] databases, were used as the sources for parent material mineralogy to allow for
20 location-specific mineralogy descriptions. In addition, a systematic protocol based on known and
21 probable silica and ferromagnesium content, spatial patterns of local and geologic settings, and
22 implied depositional mechanisms and environments was used to determine the parent material
23 acidity classifications. Therefore, steps were taken to determine accurate, location-specific
24 acidity classifications. Nonetheless, parent material in some of the plots may have been
25 misclassified.

26 To evaluate the degree to which critical load estimates could change with a
27 misclassification of parent material acidity, a simple analysis of absolute (eq/ha/yr) and
28 percentage change associated with misclassifications of parent materials was conducted, using
29 the critical loads associated with the three levels of protection ($(Bc/Al)_{crit} = 0.6, 1.2, \text{ and } 10.0$) for
30 sugar maple and red spruce. The differences between all combinations of critical loads calculated
31 with basic, intermediate, and acidic parent materials were determined, and these differences in

1 values were expressed as a percentage of the original critical load estimates (described further in
2 Appendix 5).

3 The comparisons of critical loads revealed that changes in critical load values could range
4 from 0 to 3,631 eq/ha/yr for sugar maple and 0 to 1,584 eq/ha/yr for red spruce with the
5 misclassification of parent material acidity. These ranges corresponded to percentage differences
6 ranging from 0% to 492% and 0% to 453% for sugar maple and red spruce, respectively. The
7 results also indicated that the biggest impacts of a misclassification on critical load estimates
8 would occur with an acidic parent material being misclassified as basic; the average percentage
9 changes in the estimated critical loads, in such a scenario, were 67% to 70% for sugar maple and
10 74% to 78% for red spruce, and the median percentage changes were 60% to 61% and 71% to
11 74% for the two species, respectively. In contrast, the smallest impacts on critical load estimates
12 would occur when a basic parent material was incorrectly classified as intermediate and vice
13 versa. In this scenario, the average and median percentage changes in critical load estimates were
14 only 7% to 8% and 6% to 7% for sugar maple and 5% to 6% and 4% to 5% for red spruce. Given
15 the potential significant impacts of a misclassification of parent material acidity on critical load
16 estimates, this potential source of error should be considered in the accuracy and application of
17 the critical load estimates.

18 **4.4 REFERENCES**

- 19 Arp, P.A., W. Leger, M.H. Moayeri, and J.E. Hurley. 2001. Methods for mapping forest
20 sensitivity to acid deposition for northeastern North America. *Ecosystem Health* 7(1):35–
21 47.
- 22 Bailey S.W., S.B. Horsley, R.P. Long, and R.A. Hallett. 2004. Influence of edaphic factors on
23 sugar maple nutrition and health on the Allegheny plateau. *Soil Science Society of*
24 *America Journal* 68:243–252.
- 25 Brown, L.H. 2002. *Profile of the Annual Fall Foliage Tourist in Vermont: Travel Year 2001*.
26 Report prepared for the Vermont Department of Tourism and Marketing and the Vermont
27 Tourism Data Center in association with the University of Vermont, Burlington, VT.

- 1 Bulger, A.J., B.J. Cosby, and J.R. Webb. 2000. Current, reconstructed past, and projected future
2 status of brook trout (*Salvelinus fontinalis*) streams in Virginia. *Canadian Journal of*
3 *Fisheries and Aquatic Sciences* 57:1515–1523.
- 4 Bulger, A.J., B.J. Cosby, C.A. Dolloff, K.N. Eshleman, J.R. Webb, and J.N. Galloway. 1999.
5 *SNP:FISH, Shenandoah National Park: Fish in Sensitive Habitats, Volumes I through*
6 *IV*. Project final report. Project Completion Report to the National Park Service.
7 Cooperative Agreement CA-4000-2-1007, Supplemental Agreement #2. University of
8 Virginia, Department of Environmental Sciences, Charlottesville, VA.
- 9 Chen, C.W., S.A. Gherini, J.D. Dean, R.J.M. Hudson, and R.A. Goldstein. 1984a. Development
10 and calibration of the integrated lake-watershed acidification model. Pp. 175–203 in
11 *Modeling of Total Acid Precipitation Impacts*. Edited by J.L. Schnoor. Stoneham, MA:
12 Butterworth Publishers.
- 13 Cordell, H.K., C.J. Betz, G. Green, and M. Owens. 2005. *Off-Highway Vehicle Recreation in the*
14 *United States, Regions and States: A National Report from the National Survey on*
15 *Recreation and the Environment (NSRE)*. Prepared for the U.S. Department of
16 Agriculture Forest Service, Southern Research Station, National OHV Policy and
17 Implementation Teams, Athens, GA. Available at
18 http://www.fs.fed.us/recreation/programs/ohv/OHV_final_report.pdf.
- 19 Cordell, K., B. Leeworthy, G.T. Green, C. Betz, B. Stephens. 2008. *The National Survey on*
20 *Recreation & the Environment*. Research Work Unit 4953. Pioneering Research on
21 Changing Forest Values in the South and Nation, U.S. Department of Agriculture Forest
22 Service, Southern Research Station, Athens, GA. Available at www.srs.fs.fed.us/trends.
- 23 DeHayes, D.H., P.G. Schaberg, G.J. Hawley, and G.R. Strimbeck. 1999. Acid rain impacts on
24 calcium nutrition and forest health. *Bioscience* 49(10):789–800.
- 25 Dennis, T.E., and A.J. Bulger. 1995. Condition factor and whole-body sodium concentrations in
26 a freshwater fish: Evidence for acidification stress and possible ionoregulatory
27 overcompensation. *Water, Air, and Soil Pollution* 85:377–382.

- 1 DeWalle, D. R. Swistock, B. R. 1994. Causes of episodic acidification in five Pennsylvania
2 streams on the northern Appalachian Plateau. *Water Resources Research* 30: 1955-1963.
- 3 Driscoll C. T. Van Dreason, R. 1993. Seasonal and long-term temporal patterns in the chemistry
4 of Adirondack lakes. *Water Air Soil Pollution* 67: 319-344
- 5 Driscoll, C.T., K.M. Driscoll, K.M. Roy, and M.J. Mitchell. 2003. Chemical response of lakes in
6 the Adirondacks of New York to declines in acidic deposition. *Environmental Science
7 and Technology* 37:2036–2042.
- 8 Driscoll, C. T. Postek, K. M., Kretser, E, Raynal, D. J. 1995. Long-term trends in the chemistry
9 of precipitation and lake water in the Adirondack region of New York, USA. *Water Air
10 Soil Pollution* 85: 583-588.
- 11 Driscoll, C.T., G.B. Lawrence, A.J. Bulger, T.J. Butler. C.S. Cronan, C. Eager, K.F. Lambert,
12 G.E. Likens, J.L. Stoddard, and K.C. Weathers. 2001. Acidic deposition in the
13 northeastern United States: Sources and inputs, ecosystem effects, and management
14 strategies. *Bioscience* 51(3):180–198.
- 15 Driscoll, C.T., R.M. Newton, C.P. Gubala, J.P. Baker, and S.W. Christensen. 1991. Adirondack
16 mountains. Pp. 133–202 in *Acidic Deposition and Aquatic Ecosystems: Regional Case
17 Studies*. Edited by D.F. Charles. New York, NY: Springer-Verlag.
- 18 Drohan, J. R., and W.E. Sharpe. 1997. Long-term changes in forest soil acidity in Pennsylvania,
19 U.S.A. *Water, Air, and Soil Pollution* 95:299–311.
- 20 Drohan, P.J., S.L. Stout, and G.W. Petersen. 2002. Sugar maple (*Acer saccharum* Marsh.)
21 decline during 1979-1989 in northern Pennsylvania. *Forest Ecology and Management*
22 170:1–17.
- 23 Dupont, J., T.A. Clair, C. Gagnon, D.S. Jeffries, J.S. Kahl, S.J. Nelson, and J.M. Peckenham.
24 2005. Estimation of critical loads of acidity for lakes in northeastern United States and
25 eastern Canada. *Environmental Monitoring and Assessment* 109: 275–291

- 1 Eagar, C., H. Van Miegroet, S.B. McLaughline, and N.S. Nicholas. 1996. *Evaluation of effects of*
2 *acidic deposition to terrestrial ecosystems in class I areas of the Southern Appalachians.*
3 *Technical Report. Southern Appalachian Mountains Initiative, Asheville, NC.*
- 4 Eilers J.M., G.E. Glass, K.E. Webster, and J.A. Rogalla. 1983. Hydrologic control of lake
5 susceptibility to acidification. *Canadian Journal of Fisheries and Aquatic Sciences*
6 *40:1896–1904.*
- 7 Elwood, J.W., M.J. Sale, P.R. Kaufmann, and G.F. Cada. 1991. Southern Blue Ridge Province.
8 Pp. 319–364, Chapter 11, in *Acidic Deposition and Aquatic Ecosystems: Regional Case*
9 *Studies.* Edited by D.F. Charles and S. Christie. New York, NY: Springer-Verlag.
- 10 Fenn, M.E., V.M. Perea-Estrada, L.I. de Bauer, M. Pérez-Suárez, D.R. Parker, and V.M. Cetina-
11 Alcalá. 2006. Nutrient status and plant growth effects of forest soils in the Basin of
12 Mexico. *Environmental Pollution 140(2):187–199.*
- 13 Ford, J., J.L. Stoddard, and C.F. Powers. 1993. Perspectives in environmental monitoring: an
14 introduction to the U.S. EPA Long-Term Monitoring (LTM) project. *Water, Air, and Soil*
15 *Pollution 67:247–255.*
- 16 Gran G. Determination of the equivalence point in potentiometric titrations. Part 11. *Analyst*
17 *1952;77:661-671.*
- 18 Haefele, M., R.A. Kramer, and T.P. Holmes. 1991. Estimating the total value of forest quality in
19 high-elevation spruce-fir forests. Pp. 91-96 in *Proceedings of the Conference: The*
20 *Economic Value of Wilderness.* Edited by J.M. Bowker and P.C. Reed. General Technical
21 Report. U.S. Department of Agriculture Forest Service, Southeastern Forest Experimental
22 Station, Asheville, NC.
- 23 Hall, J., B. Reynolds, S. Langan, M. Hornung, F. Kennedy, and J. Aherne. 2001. Investigating
24 the uncertainties in the simple mass balance equation for acidity critical loads for
25 terrestrial ecosystems in the United Kingdom. *Water, Air and Soil Pollution: Focus 1:*
26 *43–56.*

- 1 Hallett, R.A., S.W. Bailey, S.B. Horsley, and R.P. Long. 2006. Influence of nutrition and stress
2 on sugar maple at a regional scale. *Canadian Journal of Forest Research* 36:2235–2246.
- 3 Herlihy, A.T., Larsen, D.P., Paulsen, S.G., Urquhart, N.S., Rosenbaum, B.J., 2000. Designing a
4 spatially balanced, randomized site selection process for regional stream surveys: the
5 EMAP Mid-Atlantic Pilot Study. *Environmental Monitoring and Assessment* 63: 95–113.
- 6 Hodson, M.E., and S.J. Langan. 1999. Considerations of uncertainty in setting critical loads of
7 acidity of soils: the role of weathering rate determination. *Environmental Pollution*
8 106:73–81.
- 9 Holmes, T., and R. Kramer. 1995. An independent sample test of yea-saying and starting point
10 bias in dichotomous-choice contingent valuation. *Journal of Environmental Economics*
11 *and Management* 28:121–132.
- 12 Horsley, S.B., R.P. Long, S.W. Bailey, R.A. Hallett, and T.J. Hall. 2000. Factors associated with
13 the decline disease of sugar maple on the Allegheny Plateau. *Canadian Journal of Forest*
14 *Research* 30:1365–1378.
- 15 Hutchison, R., and C.E. Kraft. 1994. Hmong Fishing Activity and Fish Consumption. *Journal of*
16 *Great Lakes Research* 20(2):471–487.
- 17 Jenkins, D.H., J. Sullivan, and G.S. Amacher. 2002. Valuing high altitude spruce-fir forest
18 improvements: Importance of forest condition and recreation activity. *Journal of Forest*
19 *Economics* 8:77–99.
- 20 Johnson, D.W., and I.J. Fernandez. 1992. Soil-mediated effects of atmospheric deposition on
21 eastern U.S. spruce-fir forests. Pp. 235–270, Series Volume 96, in *Ecology and Decline*
22 *of Red Spruce in the Eastern United States*. Edited by C. Eagar and M.B. Adams. New
23 York, NY: Springer-Verlag.
- 24 Joslin, J.D., J.M. Kelly, and H. van Miegroet. 1992. Soil chemistry and nutrition of North
25 American spruce-fir stands: evidence for recent change. *Journal of Environmental*
26 *Quality* 21:12–30.

- 1 Kahl, J. S., Norton, S. A., Cornan, C. S. Fernandez, I. J. Bacon, K. C., Haines, T. A. 1991.
2 Maine. In Charles D. F. ed. Acidic deposition and aquatic ecosystems: regional case
3 studies, New York, NY: *Spring-Verlag*: pp. 203-235.
- 4 Kahl, J. S., Haines, T. A., Norton, S. A., Davis R. B. 1993. Recent trends in the acid-base status
5 of surface waters in Maine, USA. *Water Air Soil Pollution* 67: 281-300
- 6 Kaval, P., and J. Loomis. 2003. *Updated Outdoor Recreation Use Values with Emphasis on*
7 *National Park Recreation*. Final Report. Prepared for the National Park Service, Fort
8 Collins, CO, under Cooperative Agreement CA 1200-99-009, Project number IMDE-02-
9 0070.
- 10 Kramer, A., T. Holmes, and M. Haefel. 2003. Contingent valuation of forest ecosystem
11 protection. Pp. 303–320 in *Forests in a Market Economy*. Edited by E.O. Sills and K.L.
12 Abt. Dordrecht, The Netherlands: Kluwer Academic Publishers.
- 13 Kretser, W., J. Gallagher, and J. Nicolette. 1989. *Adirondack Lakes Study, 1984–1987: An*
14 *Evaluation of Fish Communities and Water Chemistry*. Ray Brook, NY: Adirondack
15 Lakes Survey Corporation.
- 16 Krieger, D.J. 2001. *Economic Value of Forest Ecosystem Services: A Review*. Washington, DC:
17 The Wilderness Society.
- 18 Landers, D.H., W.S. Overton, R.A. Linthurst, and D.F. Brakke. 1988. Eastern lake survey:
19 Regional estimates of lake chemistry. *Environmental Science and Technology* 22:128–
20 135.
- 21 Larsen, D.P., K.W. Thornton, N.S. Urquhart, and S.G. Paulsen. 1994. The role of sample surveys
22 for monitoring the condition of the nation's lakes. *Environmental Monitoring and*
23 *Assessment* 32:101–134.
- 24 Larsen B. P., Urquhart, N. S. (1993) A framework for assessing the sensitivity of the EMAP
25 design. In Larsen, D. P., Christie, S. J. eds. EMAP-surface waters 1991 pilot report.
26 Corvallis, OR: U.S. Environmental Protection Agency: pp. 4.1-4.37

- 1 Li, H., and S.G. McNulty. 2007. Uncertainty analysis on simple mass balance model to calculate
2 critical loads for soil acidity. *Environmental Pollution* 149:315–326.
- 3 Lien, L., G.G. Raddum, and A. Fjellheim. 1992. *Critical Loads of Acidity to Freshwater: Fish
4 and Invertebrates*. The Environmental Tolerance Levels Programme. Rep. No. 23/1992.
5 Norwegian Ministry of Environment, Oslo, Norway.
- 6 Luzadis, V.A. and E.R. Gossett. 1996. Sugar Maple. Pp. 157–166 in *Forest Trees of the
7 Northeast*. Edited by J.P. Lassoie, V.A. Luzadis, and D.W. Grover. Cooperative
8 Extension Bulletin 235. Cornell Media Services, Ithaca, NY. Available at
9 <http://maple.dnr.cornell.edu/pubs/trees.htm>.
- 10 Matuszek, J.E., and G.L. Beggs. 1988. Fish species richness in relation to lake area, pH, and
11 other abiotic factors in Ontario lakes. *Canadian Journal of Fisheries and Aquatic
12 Sciences* 45:1931–1941.
- 13 McNulty, S.G., E.C. Cohen, H. Li, and J.A. Moore-Myers. 2007. Estimates of critical acid loads
14 and exceedances for forest soils across the conterminous United States. *Environmental
15 Pollution* 149:281–292.
- 16 MEA (Millennium Ecosystem Assessment). 2005. *Ecosystems and Human Well-being: Wetlands
17 and Water. Synthesis. A Report of the Millennium Ecosystem Assessment*. Washington,
18 DC: World Resources Institute.
- 19 Murdoch, P. S. Stoddard, J. L. (1993) Chemical characteristics and temporal trends in eight
20 streams of the Catskill Mountains, New York. *Water Air Soil Pollution* 67: 367-395.
- 21 NAPAP (National Acid Precipitation Assessment Program). 2005. *National Acid Precipitation
22 Assessment Program Report to Congress: An Integrated Assessment*. National Acid
23 Precipitation Assessment Program, Washington, DC.
- 24 NAPAP (National Acid Precipitation Assessment Program). 1998. *NAPAP Biennial Report to
25 Congress: An Integrated Assessment*. National Science and Technology Council,
26 Committee on Environment and Natural Resources, Silver Spring, MD.

- 1 NASS (National Agricultural Statistics Service). 2008. *Maple Syrup – June 12, 2008: Maple*
2 *Syrup Production Up 30 Percent Nationwide*. U.S. Department of Agriculture, National
3 Agricultural Statistics Service, New England Agricultural Statistics, Concord, NH.
- 4 NEG/ECP Forest Mapping Group (Conference of New England Governors and Eastern
5 Canadian Premiers Forest Mapping Group). 2001. *Protocol for Assessment and Mapping*
6 *of Forest Sensitivity to Atmospheric S and N Deposition: Acid Rain Action Plan – Action*
7 *Item 4: Forest Mapping Research Project*. Prepared by NEG/ECP Forest Mapping
8 Group. Available at
9 [http://www.nrs.fs.fed.us/clean_air_water/clean_water/critical_loads/local-](http://www.nrs.fs.fed.us/clean_air_water/clean_water/critical_loads/local-resources/docs/NEGECP_Forest_Sensitivity_Protocol_5_21_04.pdf)
10 [resources/docs/NEGECP_Forest_Sensitivity_Protocol_5_21_04.pdf](http://www.nrs.fs.fed.us/clean_air_water/clean_water/critical_loads/local-resources/docs/NEGECP_Forest_Sensitivity_Protocol_5_21_04.pdf).
- 11 Webb, R. 2004. *Effects of Acidic Deposition on Aquatic Resources in the Central Appalachian*
12 *Mountains*. University of Virginia, Department of Environmental Sciences.
13 <http://swas.evsc.virginia.edu>
- 14 Ouimet, R., P.A. Arp, S.A. Watmough, J. Aherne, and I. DeMerchant. 2006. Determination and
15 mapping critical loads of acidity and exceedances for upland forest soils in Eastern
16 Canada. *Water, Air and Soil Pollution* 172:57–66.
- 17 Pardo, L.H., and C.T. Driscoll. 1996. Critical loads for nitrogen deposition: case studies at two
18 northern hardwood forests. *Water, Air, and Soil Pollution* 89:105–128.
- 19 Pardo, L.H., and N. Duarte. 2007. *Assessment of Effects of Acidic Deposition on Forested*
20 *Ecosystems in Great Smoky Mountains National Park using Critical Loads for Sulfur and*
21 *Nitrogen*. U.S. Department of Agriculture, Forest Service. Prepared for Tennessee Valley
22 Authority, S. Burlington, VT.
- 23 Peterson, D.E., M.S. Kanarek, M.A. Kuykendall, J.M. Diedrich, H.A. Anderson, P.L.
24 Remington, and T.B. Sheffy. 1994. Fish Consumption Patterns and Blood Mercury
25 Levels in Wisconsin Chippewa Indians. *Archives of Environmental Health* 49(1):53–58.
- 26 Rago, P.J., and J.G. Wiener. 1986. Does pH affect fish species richness when lake area is
27 considered? *Transactions of the American Fisheries Society* 115:438–447.

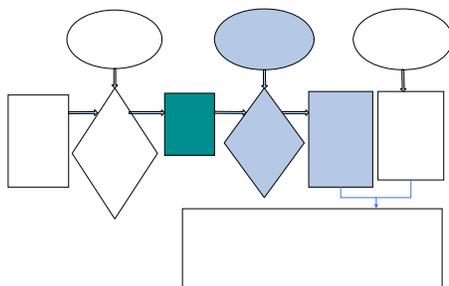
- 1 Reuss, J.O., and D.W. Johnson. 1985. Effect of soil processes on the acidification of water by
2 acid deposition. *Journal of Environmental Quality* 14:26–31.
- 3 Schreck, C.B. 1982. Stress and compensation in teleostean fishes: response to social and physical
4 factors. Pp. 295–321 in *Stress and Fish*. Edited by A.D. Pickering. New York, NY:
5 Academic Press.
- 6 Schreck, C.B. 1981. Stress and rearing of salmonids. *Aquaculture* 28:241–249.
- 7 Spencer, D.M., and D.F. Holecek. 2007. Basic characteristics of the fall tourism market. *Tourism*
8 *Management* 28:491–504.
- 9 St. Clair, S.B., J.E. Carlson, and J.P. Lynch. 2005. Evidence for oxidative stress in sugar maple
10 stands growing on acidic, nutrient imbalanced forest soils. *Oecologia* 145:258–369.
- 11 Stoddard, J., J.S. Kahl, F.A. Deviney, D.R. DeWalle, C.T. Driscoll, A.T. Herlihy, J.H. Kellogg,
12 P.S. Murdoch, J.R. Webb, and K.E. Webster. 2003. *Response of Surface Water Chemistry*
13 *to the Clean Air Act Amendments of 1990*. EPA 620/R-03.001. U.S. Environmental
14 Protection Agency, Office of Research and Development, National Health and
15 Environmental Effects Research Laboratory, Research Triangle Park, NC.
- 16 Stoddard, J.L., C.T. Driscoll, J.S. Kahl, J.H. Kellogg. 1998. A regional analysis of lake
17 acidification trends for the northeastern U.S., 1982–1994. *Environmental Monitoring and*
18 *Assessment* 51:399–413.
- 19 Stoddard, J. L., Urquhart, N. S. Newell, A. D. Kugler, D. (1996) The Temporally Integrated
20 Monitoring of Ecosystems (TIME) project cesign. 2, Detection of regional acidification
21 trends. *Water Resources*. 32: 2529-2538
- 22 Stoddard, J.L. 1990. *Plan for Converting the NAPAP Aquatic Effects Long-Term Monitoring*
23 *(LTM) Project to the Temporally Integrated Monitoring of Ecosystems (TIME) Project*.
24 International report. U.S. Environmental Protection Agency, Corvallis, OR.
- 25 Sullivan, T.J., C.T. Driscoll, B.J. Cosby, I.J. Fernandez, A.T. Herlihy, J. Zhai, R. Stemberger,
26 K.U. Snyder, J.W. Sutherland, S.A. Nierzwicki-Bauer, C.W. Boylen, T.C. McDonnell,

- 1 and N.A. Nowicki. 2006. *Assessment of the Extent to which Intensively-Studied Lakes are*
2 *Representative of the Adirondack Mountain Region*. Final report. New York State Energy
3 Research and Development Authority (NYSERDA), Albany, NY. Available at
4 <http://nysl.nysed.gov/uhtbin/cgisirsi/Qcwd6NzFby/NYSL/138650099/8/4298474>
5 (accessed November 1, 2007).
- 6 Sverdrup, H., and W. de Vries. 1994. Calculating critical loads for acidity with the simple mass
7 balance equation. *Water, Air, and Soil Pollution* 72:143–162.
- 8 Sverdrup, H., and P. Warfvinge. 1993a. *The effect of soil acidification on the growth of trees,*
9 *grass and herbs as expressed by the (Ca+ Mg+ K)/Al ratio*. Reports in Ecology and
10 Environmental Engineering 2. Lund University, Department of Chemical Engineering,
11 Lund, Sweden.
- 12 UNECE (United Nations Economic Commission for Europe). 2004. *Manual on Methodologies*
13 *and Criteria for Modeling and Mapping Critical Loads and Levels and Air Pollution*
14 *Effects, Risks, and Trends*. Convention on Long-Range Transboundary Air Pollution,
15 Geneva, Switzerland. Available at <http://www.icpmapping.org> (accessed August 16,
16 2006).
- 17 Urquhart et al., 1998
- 18 USDA-NRCS (United States Department of Agriculture-Natural Resources Conservation
19 Service). 2008. *Soil Survey Geographic (SSURGO) Database*. U.S. Department of
20 Agriculture, Natural Resources Conservation Service, Fort Worth, TX. Available at
21 <http://datagateway.nrcs.usda.gov>.
- 22 U.S. EPA (Environmental Protection Agency). 2008. *Integrated Science Assessment (ISA) for*
23 *Oxides of Nitrogen and Sulfur–Ecological Criteria (Final Report)*. EPA/600/R-
24 08/082F. U.S. Environmental Protection Agency, National Center for Environmental
25 Assessment–RTP Division, Office of Research and Development, Research Triangle
26 Park, NC. Available at <http://cfpub.epa.gov/ncea/cfm/recordisplay.cfm?deid=201485>.

- 1 U.S. EPA (Environmental Protection Agency). 2005. *EPA Advisory Council on Plans for*
2 *Ecological Effects Analysis in the Analytical Plan for EPA’s Second Prospective Analysis*
3 *– Benefits and Costs of the Clean Air Act, 1990-2020*. EPA-COUNCIL-ADV-05-001.
4 U.S. Environmental Protection Agency, Washington, DC. June 23.
- 5 U.S. EPA (Environmental Protection Agency). 2000. *Guidelines for Preparing Economic*
6 *Analyses*. Washington, DC: National Center for Environmental Economics.
- 7 U.S. EPA (Environmental Protection Agency). 1995. *Review of the national ambient air quality*
8 *standards for nitrogen dioxide: Assessment of scientific and technical information*.
9 OAQPS staff paper. EPA-452/R-95-005. U.S. Environmental Protection Agency, Office
10 of Air Quality Planning and Standards, Research Triangle Park, NC.
- 11 USFS (U.S. Forest Service). 2006. *Forest Inventory and Analysis National Program: Forest*
12 *Inventory Data Online*. Online database. U.S. Department of Agriculture Forest Service,
13 Forest Inventory and Analysis, Arlington, VA. Available at <http://fia.fs.fed.us/tools-data>.
- 14 USFS (U.S. Forest Service). 2002a. *Forest Inventory and Analysis National Program: Area of*
15 *Forest Land, 2002*. U.S. Department of Agriculture, Forest Service, Forest Inventory and
16 Analysis National Program, Arlington, VA. Available at [http://fia.fs.fed.us/tools-](http://fia.fs.fed.us/tools-data/maps/descr/for_land.asp)
17 [data/maps/descr/for_land.asp](http://fia.fs.fed.us/tools-data/maps/descr/for_land.asp) (accessed March 3, 2009).
- 18 USFS (U.S. Forest Service). 2002b. *Forest Inventory and Analysis National Program: Area of*
19 *Timberland, 2002*. U.S. Department of Agriculture, Forest Service, Forest Inventory and
20 Analysis National Program, Arlington, VA. Available at [http://fia.fs.fed.us/tools-](http://fia.fs.fed.us/tools-data/maps/descr/tim_land.asp)
21 [data/maps/descr/tim_land.asp](http://fia.fs.fed.us/tools-data/maps/descr/tim_land.asp) (accessed March3, 2009).
- 22 U.S. FWS (Fish and Wildlife Service) and U.S. Census Bureau. 2007. *2006 National Survey of*
23 *Fishing, Hunting, and Wildlife-Associated Recreation*. FHW/06-NAT. U.S. Department
24 of the Interior, U.S. Fish and Wildlife Service, Washington, DC, and U.S. Department of
25 Commerce, U.S. Census Bureau, Washington, DC.
- 26 USGS (U.S. Geological Survey). 2009. *State Geological Map Compilation*. U.S. Geological
27 Survey, Mineral Resources On-Line Spatial Data. U.S. Department of the Interior, U.S.

- 1 Geological Survey, Reston, VA. Available at <http://tin.er.usgs.gov/geology/state>
2 (accessed January 28, 2009).
- 3 Van der Salm, C., and W. de Vries. 2001. A review of the calculation procedure for critical acid
4 loads for terrestrial ecosystems. *The Science of the Total Environment* 271:11–25.
- 5 Warby, R.A.F., C.E. Johnson, and C.T. Driscoll. 2009. Continuing acidification of organic soils
6 across the northeastern USA: 1984-2001. *Soil Science Society of America Journal*
7 73:274–284.
- 8 Watmough, S., J. Aherne, P. Arp, I. DeMerchant, and R. Ouimet. 2006. Canadian experiences in
9 development of critical loads for sulphur and nitrogen. Pp. 33–38 in *Monitoring Science*
10 *and Technology Symposium: Unifying Knowledge for Sustainability in the Western*
11 *Hemisphere Proceedings RMRS-P-42CD*. Edited by C. Aguirre-Bravo, P.J. Pellicane,
12 D.P. Burns, and S. Draggan. U.S. Department of Agriculture, Forest Service, Rocky
13 Mountain Research Station, Fort Collins, CO.
- 14 Watmough, S.A., J. Aherne, and P.J. Dillion. 2004. *Critical Loads Ontario: Relating Exceedance*
15 *of the Critical Load with Biological Effects at Ontario Forests*. Report 2. Environmental
16 and Resource Studies, Trent University, ON, Canada.
- 17 Webb, J.R., B.J. Cosby, F.A. Deviney, J.N. Galloway, S.W. Maben, and A.J. Bulger. 2004. Are
18 brook trout streams in western Virginia and Shenandoah National Park recovering from
19 acidification? *Environmental Science and Technology* 38:4091–4096.
- 20 Webb J.R., Cosby B.J., Galloway J.N., and Hornberger G.M. 1989. Acidification of native brook
21 trout streams in Virginia. *Water Resources Research* 25:1367–1377.
- 22 Webster, E., and D. Mackay, 2003. *Defining Uncertainty and Variability in Environmental Fate*
23 *Models*. CEMC Report No. 200301. Canadian Environmental Modelling Centre, Trent
24 University, Peterborough, ON, Canada. Available at
25 www.trentu.ca/cemc/CEMC200301.pdf.

- 1 Webster, K.L., I.F. Creed, N.S. Nicholas, H.V. Miegroet. 2004. Exploring interactions between
2 pollutant emissions and climatic variability in growth of red spruce in the Great Smoky
3 Mountains National Park. *Water, Air, and Soil Pollution* 159:225–248.
- 4 Webster, K. E., Brezonik, P. L., Holdhsen, B. J. 1993. Temporal trends in low alkalinity lakes of
5 the upper Midwest (1983-1989). *Water Air Soil Pollution* 67: 397-414.
- 6 Wedemeyer, G.A., B.A. Barton, and D.J. MeLeay. 1990. Stress and acclimation. Pp. 178–198 in
7 *Methods for Fish Biology*. Edited by C.B. Schreck and P.B. Moyle. American Fisheries
8 Society, Bethesda, MD.

1
23

5.0 NUTRIENT ENRICHMENT

4

5.1 SCIENCE OVERVIEW

6 Nitrogen and sulfur enrichment represents a
 8 continuum of effects that can be characterized as positive or
 10 negative, depending on the selected ecological endpoint,
 12 location, and baseline conditions of an ecosystem. Nutrient
 14 enrichment describes a condition where an increase in a
 16 nutrient, such as nitrogen, may result in an imbalance in
 18 ecological stoichiometry, causing effects on ecological
 19 processes, structure, and function. Organisms in their natural environment are commonly adapted
 20 to a specific regime of nutrient availability (Sterner and Elser, 2002). Some organisms may at
 21 first respond positively to an initial increase in nutrients, exhibiting a fertilized increase in
 22 growth. However, as the nutrient load continues to rise, the imbalance can have negative effects
 23 in the organism's response or the invasion of new organisms that benefit from increased
 24 nutrients. In general, ecosystems that are most responsive to nutrient enrichment from
 25 atmospheric nitrogen deposition are those that receive high levels of deposition relative to
 26 nonanthropogenic nitrogen loading, those that are nitrogen-limited, or those that contain species
 27 that have evolved in nutrient-poor environments (U.S. EPA, 2008, Section 3.3). Nutrient
 28 enrichment in ecosystems may alter the native terrestrial species composition (e.g., species shift
 29 from wildflower meadows to shrubs) and can result in eutrophication in aquatic systems (see
 30 Section 3.3 of the *Integrated Science Assessment (ISA) for Oxides of Nitrogen and Sulfur–*
 31 *Ecological Criteria (Final Report) (ISA)* (U.S. EPA, 2008).

Nutrient enrichment is an increase in a nutrient, such as nitrogen, that may result in an imbalance in ecological stoichiometry, causing effects on processes, structure, and function. Organisms in their natural environment are commonly adapted to a specific regime of nutrient availability (Sterner and Elser, 2002).

1 Both aquatic and terrestrial effects of nutrient enrichment have been studied, and nitrogen
2 enrichment is highlighted in this chapter. (Sulfur enrichment is discussed in Chapter 6.) For each
3 effect, information is presented on the following:

- 4 ▪ Ecological indicators, ecological responses, and ecosystem services
- 5 ▪ Characteristics of areas sensitive to nutrient enrichment
- 6 ▪ Selection of case study area(s)
- 7 ▪ Current conditions in case study areas
- 8 ▪ The ability to extrapolate case study findings to larger regions
- 9 ▪ Current conditions for larger regions (based on extrapolation)
- 10 ▪ Ecological effect functions
- 11 ▪ Uncertainty and variability associated with the case study analyses.

12 Case studies on aquatic nutrient enrichment and terrestrial nutrient enrichment were
13 performed as part of this Risk and Exposure Assessment (Appendices 6 and 7, respectively) to
14 aid in determining whether a link can be established between deposition of nitrogen oxides
15 (NO_x) (and/or total reactive nitrogen) and ecosystem response, as well as the impact of total
16 reactive nitrogen deposition relative to NO_x deposition. These case studies are also intended to
17 test whether area-based risk and exposure assessments are a suitable method for predicting
18 nutrient enrichment effects on other ecosystems and geographic regions. The studies facilitate
19 extrapolation of impacts from smaller-scale that are representative of sensitive areas to similar
20 ecosystems across the country.

21 **5.1.1 Aquatic Nutrient Enrichment**

22 Nutrient enrichment can result in eutrophication of aquatic systems (U.S. EPA, 2008,
23 Section 3.3). Eutrophication is the process whereby a body of water becomes overenriched in
24 nutrients, resulting in increased productivity (e.g., of algae or aquatic plants). As productivity
25 increases, dissolved oxygen levels in the waterbody may decrease and lead to hypoxia (i.e., low
26 dissolved oxygen levels). Total reactive nitrogen (Nr) can promote eutrophication in inland
27 freshwater, estuarine, and coastal marine ecosystems. Eutrophication ultimately reduces
28 biodiversity because of the lack of available oxygen needed for the survival of many aquatic
29 plants and animals. The ISA concluded that there is sufficient evidence to infer a causal

1 relationship between nitrogen deposition and the biogeochemical cycling of nitrogen in estuaries
2 and coastal marine waters. Atmospheric nitrogen deposition is not the sole source of nitrogen
3 loading to estuaries, and it is unknown if atmospheric deposition alone is sufficient to cause
4 eutrophication. However, the contribution of atmospheric nitrogen deposition to total nitrogen
5 load is calculated for some estuaries and can be >40%. In general, estuaries tend to be nitrogen-
6 limited, and many currently receive high levels of nitrogen input from human activities to cause
7 eutrophication. Because ecosystems may respond differently to enrichment, it is necessary to
8 first perform risk and exposure assessments unique to the effect and ecosystem type. Appendix 6
9 presents a case study on two river basins and their estuaries: the Potomac River/Potomac Estuary
10 and the Neuse River/Neuse River Estuary, and Section 5.2 summarizes the science,
11 methodologies, and findings of the Aquatic Nutrient Enrichment Case Study.

12 **5.1.2 Terrestrial Nutrient Enrichment**

13 The ISA (U.S. EPA, 2008, Section 3.3) documented the current understanding of nutrient
14 enrichment effects on terrestrial ecosystems and concluded that there is sufficient information to
15 infer a causal relationship between atmospheric nitrogen deposition and biogeochemical cycling
16 and fluxes of nitrogen in terrestrial systems. The ISA also concluded that there is a causal
17 relationship between atmospheric nitrogen deposition and changes in species richness, species
18 composition, and biodiversity in terrestrial systems. These conclusions are based on an extensive
19 literature review, which is summarized in Table 4-4 of the ISA. The research involves both
20 observational and experimental (nitrogen-addition) projects and includes alpine ecosystems,
21 grasslands (including arid and semiarid ecosystems), forests, and deserts. This extensive
22 documentation was used to assist in the selection of the case study areas for this Risk and
23 Exposure Assessment and to identify and compare ecological benchmarks of different
24 ecosystems. Appendix 7 presents the case study report for two ecosystems: California coastal
25 sage scrub (CSS) and San Bernardino Mountains mixed conifer forest (MCF). Section 5.3
26 summarizes the Terrestrial Nutrient Enrichment Case Study.

27 **5.2 AQUATIC NUTRIENT ENRICHMENT**

28 Aquatic nutrient enrichment is described in the ISA (U.S. EPA, 2008, Section 3.3) for
29 both freshwater and coastal marine and estuarine systems. In nitrogen-limited freshwater aquatic
30 systems, atmospheric inputs of nitrogen increase productivity and alter biological communities,

1 especially phytoplankton. A freshwater lake or stream must be nitrogen-limited to be sensitive to
2 nitrogen-mediated eutrophication. There are many examples of fresh waters that are nitrogen-
3 limited or nitrogen and phosphorus co-limited (e.g., Baron, 2006; Bergström and Jansson, 2006;
4 Bergström et al., 2005; Elser et al., 1990; Fenn et al., 2003; Tank and Dodds, 2003). In a meta-
5 analysis that included 653 datasets, Elser et al. (2007) found that nitrogen limitation occurred as
6 frequently as phosphorus limitation in freshwater ecosystems. Recently, a comprehensive study
7 (Bergström and Jansson, 2006) of available data from the northern hemisphere survey of lakes
8 along gradients of nitrogen deposition showed increased inorganic nitrogen concentrations and
9 productivity to be correlated with atmospheric nitrogen deposition, leading to the conclusion that
10 the results are evidence of nitrogen limitation in lakes with low ambient inputs of nitrogen and
11 increased nitrogen concentration in lakes receiving nitrogen solely from atmospheric nitrogen
12 deposition (Bergström and Jansson, 2006).

13 In coastal marine ecosystems, the nutrients most commonly associated with
14 phytoplankton growth are nitrogen, phosphorus, and silicon. Interactions among the supplies of
15 these nutrients can affect phytoplankton species composition in ways that might affect ecosystem
16 function (Paerl et al., 2001a; Riegman, 1992). The relative proportions of these nutrients are
17 important determinants of primary production, food web structure, and energy flow through the
18 ecosystem (Dortch and Whitley, 1992; Justic et al., 1995a; Justic et al., 1995b; Turner et al.,
19 1998).

20 There is strong scientific consensus that nitrogen is the principal cause of coastal
21 eutrophication in the United States (NRC, 2000). On average, human activity has likely
22 contributed to a six-fold increase in the nitrogen flux to U.S. coastal waters, and nitrogen now
23 represents the most significant coastal pollution problem (Howarth et al., 2002b; Howarth and
24 Marino, 2006). Atmospheric deposition is responsible for a portion of the nitrogen input
25 (Howarth et al., 2002a).

26 Estuaries and coastal waters tend to be nitrogen-limited and are, therefore, inherently
27 sensitive to increased nitrogen loading (D'Elia et al., 1986; Howarth and Marino, 2006). There is
28 a scientific consensus that nitrogen-driven eutrophication in shallow estuaries has increased over
29 the past several decades and that the environmental degradation of coastal ecosystems is now a
30 widespread occurrence (Paerl et al., 2001a). For example, the frequency of phytoplankton
31 blooms and the extent and severity of hypoxia have increased in the Chesapeake Bay (Officer et

1 al., 1984) and Pamlico estuaries in North Carolina (Paerl et al., 1998) and along the continental
2 shelf adjacent to the Mississippi and Atchafalaya rivers' discharges to the Gulf of Mexico (Eadie
3 et al., 1994). It is partly because many estuaries and near-coastal marine waters are degraded by
4 nutrient enrichment that they are highly sensitive to potential negative impacts from nitrogen
5 addition from atmospheric deposition.

6 The Aquatic Nutrient Enrichment Case Study for this Risk and Exposure Assessment
7 (Appendix 6) focuses on two estuarine systems—the Potomac Estuary and the Neuse River
8 Estuary. The ecological indicator selected, risk and exposure assessment methodology, and
9 findings for each system are described in this section.

10 **5.2.1 Ecological Indicators, Ecological Responses, and Ecosystem Services**

11 **5.2.1.1 Indicators**

12 **Overview.** Nitrogen is an essential nutrient for estuarine and marine ecosystem fertility; a
13 key nutrient in the primary production of aquatic vegetation; and is often the algal growth-
14 limiting nutrient (U.S. EPA, 2008, Section 3.3.5.3). Excessive nitrogen contributions increase
15 primary productivity excessively and, in turn, cause habitat degradation, algal blooms, toxicity,
16 hypoxia, anoxia, fish kills, and reductions in biodiversity (Paerl, 2002). To evaluate these
17 impacts, five biological indicators were used in the recent national assessment of estuary trophic
18 condition: chlorophyll *a*, macroalgae, dissolved oxygen, nuisance/toxic algal blooms, and
19 submerged aquatic vegetation (SAV) (Bricker et al., 2007). **Figure 5.2-1**, excerpted from the
20 National Oceanic and Atmospheric Administration's (NOAA's) National Estuarine
21 Eutrophication Assessment (NEEA) Update, provides a brief description of each of the
22 indicators. For greater detail on each of the indicators, refer to the ISA (U.S. EPA, 2008, Section
23 3.3) and the NEEA Update (Bricker et al., 2007).

Primary symptoms		Description
	Chlorophyll <i>a</i> (Phytoplankton)	A measure used to indicate the amount of microscopic algae (phytoplankton) growing in a water body. High concentrations can lead to low dissolved oxygen levels as a result of decomposition.
	Macroalgal blooms	Large algae commonly referred to as “seaweed.” Blooms can cause losses of submerged aquatic vegetation by blocking sunlight. Additionally, blooms may smother immobile shellfish, corals, or other habitat. The unsightly nature of some blooms may impact tourism due to the declining value of swimming, fishing, and boating.
Secondary symptoms		Description
	Dissolved oxygen	Low dissolved oxygen is a eutrophic symptom because it occurs as a result of decomposing organic matter (from dense algal blooms), which sinks to the bottom and uses oxygen during decay. Low dissolved oxygen can cause fish kills, habitat loss, and degraded aesthetic values, resulting in the loss of tourism and recreational water use.
	Submerged aquatic vegetation	Loss of submerged aquatic vegetation (SAV) occurs when dense algal blooms caused by excess nutrient additions (and absence of grazers) decrease water clarity and light penetration. Turbidity caused by other factors (e.g., wave energy, color) similarly affects SAV. The loss of SAV can have negative effects on an estuary’s functionality and may impact some fisheries due to loss of a critical nursery habitat.
	Nuisance/toxic blooms	Thought to be caused by a change in the natural mixture of nutrients that occurs when nutrient inputs increase over a long period of time. These blooms may release toxins that kill fish and shellfish. Human health problems may also occur due to the consumption of contaminated shellfish or from inhalation of airborne toxins. Many nuisance/toxic blooms occur naturally, some are advected into estuaries from the ocean; the role of nutrient enrichment is unclear.

1
2 **Figure 5.2-1.** Descriptions of the five eutrophication indicators used in the NEEA
3 (Bricker et al., 2007).

4 **Selection of an Ecological Indicator.** After examining several estuarine assessment
5 options, the most comprehensive evaluation technique that could be applied on a wide scale was
6 determined to be an assessment of eutrophication as conducted in NOAA’s NEEA. The NEEA
7 Program defined and developed a Pressure-State-Response framework to assess the potential for
8 eutrophication. This assessment framework has been titled the Assessment of Estuarine Trophic
9 Status Eutrophication Index and is commonly referred to as ASSETS EI (Bricker et al., 2007).
10 The “pressure” is the nitrogen, the “state” is the current eutrophic condition, and the “response”
11 would be the change in the state of the system. ASSETS EI is an estimation of the likelihood that
12 the estuary is experiencing eutrophication or will experience eutrophication in the future based
13 on the five indicators described above. The ASSETS EI served as the ecological indicator for the
14 Aquatic Nutrient Enrichment Case Study.

15 The ASSETS EI incorporates indirect deposition over the watershed (i.e., deposition to
16 terrestrial systems which, in turn, may be transported to aquatic systems) through the evaluation
17 of nitrogen loading to the estuary. This was achieved by inputting 2002 Community Multiscale
18 Air Quality (CMAQ)–modeled and National Atmospheric Deposition Program (NADP)–

1 monitored data (see Chapter 3) to the U.S. Geological Survey's (USGS's) SPATIally Referenced
2 Regressions on Watershed attributes (SPARROW) model. The combination of SPARROW
3 modeling and the ASSETS EI (Appendix 6, Figure 2.2-1) provides a sound basis for conducting
4 an eutrophication assessment.

5 **ASSETS EI.** The ASSETS EI (a Pressure-State-Response framework) is categorical,
6 where each of three indices produces a score. The three scores are combined, and the overall
7 score (the ASSETS EI) represents the estuary's health. The indices are as follows:

- 8 ■ **Influencing Factors/Overall Human Influence (OHI).** The physical, hydrologic, and
9 anthropogenic factors that characterize the susceptibility of the estuary to the influences of
10 nutrient inputs (also quantified as part of the index) and eutrophication.
- 11 ■ **Overall Eutrophic Condition (OEC).** An estimate of current eutrophic conditions
12 derived from data for five symptoms known to be linked to eutrophication.
- 13 ■ **Determined Future Outlook (DFO).** A qualitative measure of expected changes in the
14 system.

15 (See Figures 2.2-6 and 2.2-8 in Appendix 6 for the ASSETS EI approach to assessing
16 OEC and DFO.)

17 The ASSETS EI scores fall into one of six categories: High, Good, Moderate, Poor, Bad,
18 or Unknown. These ratings can be summarized as follows (Bricker et al., 2007):

- 19 ■ **High:** Low pressure, low eutrophic condition, and any expected improvement or no
20 future change in eutrophic condition
- 21 ■ **Good:** Low to moderate pressure, low to moderate-low eutrophic condition, and any
22 expected future change in condition
- 23 ■ **Moderate:** Any pressure, moderate-low to moderate-high eutrophic condition, and any
24 expected future change in eutrophic condition
- 25 ■ **Poor:** Moderate-low to high pressure, moderate to moderate-high eutrophic condition,
26 and any expected future change in condition
- 27 ■ **Bad:** Moderate to high pressure, moderate-high to high eutrophic condition, and any
28 expected future change in eutrophic condition
- 29 ■ **Unknown:** Insufficient data for analysis.

1 NOAA’s ASSETS EI method was first reported in 1999. Since that time, it has been used
2 in several assessments across the country and internationally, and it has undergone revision and
3 validation (Bricker et al., 1999, 2003, 2007; Ferreira et al., 2007; Whitall et al., 2007).

4 **5.2.1.2 Assessments of Ecological Responses Using SPARROW and ASSETS EI**

5 To assess ecological response, the SPARROW output serves as the nitrogen load for the
6 calculation of the OHI index in the ASSETS EI. In this case study, a complete analysis from
7 atmospheric deposition load to the ASSETS EI ecological endpoint requires the following:

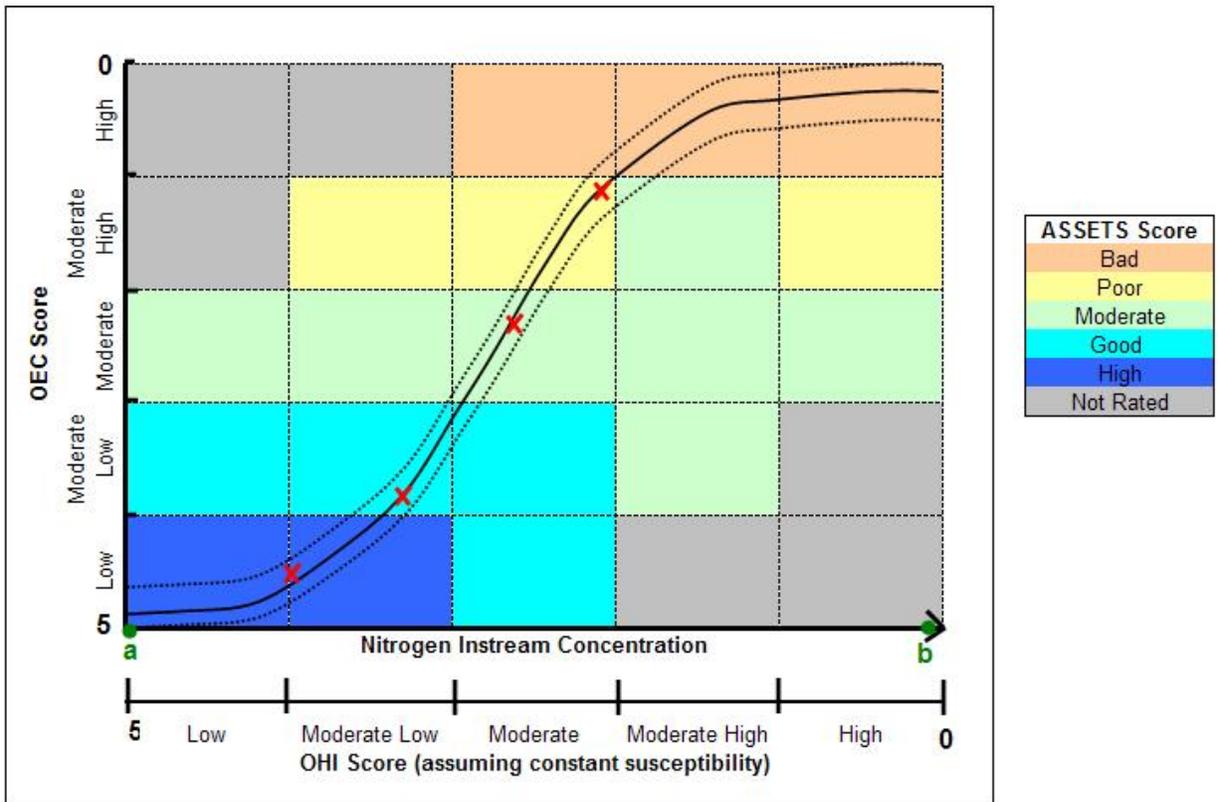
- 8 ■ An assessment of the relative changes in the deposition load
- 9 ■ The resulting instream nitrogen load to the estuary
- 10 ■ The change in the ASSETS EI.

11 Because an iterative assessment of changing nitrogen loads to predict ASSETS EIs has
12 not been undertaken previously, a process to link the SPARROW model to the ASSETS EI was
13 developed and used (See Appendix 6, Section 2.2.3).

14 A series of response curves was created to relate nitrogen inputs to ecosystem responses
15 in the watershed and estuary. First, the SPARROW model was used to predict the total nitrogen
16 loads at the outlet of the watershed that result from changes in the total nitrogen atmospheric
17 deposition loads (i.e., changes in the ambient air NO_x concentrations and subsequent deposition
18 that result from any new standard-setting scenarios). Second, a response curve was plotted for
19 the ASSETS EI based on the OHI and OEC index scores (Appendix 6, Section 2.2.2), which are
20 functions of total nitrogen load to the estuary. Bricker et al. (2007) noted that the shape of the
21 response curve would vary depending on the susceptibility of the system. Therefore, if the
22 susceptibility is known and held constant, a curve can be created.

23 It is possible to combine all the OEC, OHI, and DFO index scores with the ASSETS EI
24 into a single response curve when the susceptibility rating and DFO index score are held
25 constant. The DFO index score may be held constant when alternative effects levels are being
26 evaluated based on a current condition scenario. The susceptibility rating is based on physical
27 and hydrological conditions, which are unlikely to change. For example, **Figure 5.2-2** highlights
28 this combination of scores where the susceptibility rating is “High” and the DFO index score is
29 set at “Improve.” Additionally, by holding the susceptibility constant, the OHI index score
30 becomes a function of the instream nitrogen concentration. This is evident in the double x-axis.

- 1 The state response is the OEC index score along the y-axis. Underlying these combinations of
- 2 OHI and OEC index scores is the ASSETS EI.



3
4 **Figure 5.2-2.** ASSETS EI response curve.

5 Within the analysis space created by both the OHI and OEC index scores, the axes are
6 limited to the scores of zero (actually categorized as one in the NEEA Update) to five, but the
7 corresponding instream nitrogen concentrations must be determined separately. Point “a”
8 represents the background nitrogen concentration that would occur in the system with no
9 anthropogenic inputs (assuming the system is not naturally eutrophic) or with the system at a
10 pristine state. In almost all cases, this value will be unknown because of the extent to which
11 anthropogenic inputs have influenced the nation’s ecosystems. A lower bound and upper bound
12 on this value were specified between which the algorithm randomly selects a different realization
13 for each iteration. The upper bound of the instream total nitrogen concentration (TN_s), Point “b,”
14 is the maximum nitrogen concentration at which the system is nitrogen-limited; above this point,
15 the nitrogen inputs to the system no longer affect the eutrophication condition. Again, because of

1 natural variations, a constant value is unknown, and upper and lower bounds of the value must be
2 specified for uncertainty analyses.

3 The creation of the two response curves enables working backward from the ecological
4 endpoint to the source of the impairment; in this case, from the ASSETS EI to the atmospheric
5 deposition loading of oxidized nitrogen. Specifically, the analysis in this case study sought to
6 determine the change in oxidized nitrogen load required to improve the ASSETS EI by one, two,
7 and three categories from its current level set in the 2002 current condition analysis.

8 **5.2.1.3 Ecosystem Services**

9 **Provisioning Services**

10 Estuaries in the eastern United States are an important source of food production, in
11 particular fish and shellfish production. The estuaries are capable of supporting large stocks of
12 resident commercial species, and they serve as the breeding grounds and interim habitat for
13 several migratory species.

14 To provide an indication of the magnitude of provisioning services associated with
15 coastal fisheries, from 2005 to 2007, the average value of total catch was \$1.5 billion per year in
16 15 East Coast states. It is not known, however, what percentage of this value is directly
17 attributable to or dependent upon the estuaries in these states. **Table 5.2-1** focuses specifically on
18 commercial landings in Maryland and Virginia in 2007 and reports values for the main
19 commercial species in these states. Although these values also include fish caught outside of the
20 Chesapeake Bay, the values for two key species—blue crab and striped bass—are predominantly
21 from the estuary itself. These data indicate that blue crab landings in 2007 totaled nearly \$44
22 million in the Chesapeake Bay. The value of striped bass and menhaden totaled about \$9 million
23 and \$25 million, respectively.

24 To most accurately assess how eutrophication in East Coast estuaries is related to the
25 long-term provisioning services from their fishery resources requires bioeconomic models (i.e.,
26 models that combine biological models of fish population dynamics with economic models
27 describing fish harvesting and consumption decisions). In most cases, these models address the
28 dynamic feedback effects between fish stocks and harvesting behavior, and they characterize
29 conditions for a “steady-state” equilibrium, where stocks and harvest levels are stabilized and
30 sustainable over time.

1 Section 5.2 describes one bioeconomic model linking blue crab harvests to nutrient loads
 2 in the Neuse River Estuary, and it applies the model to estimate how reductions in nitrogen loads
 3 to the estuary would affect the societal value of future blue crab harvests. In practice, however,
 4 very few other studies have developed empirical bioeconomic models to estimate how changes
 5 in environmental quality affect fish harvests and the value of these services (Knowler, 2002).
 6 One exception is Kahn and Kemp (1985), which estimated a bioeconomic model of commercial
 7 and recreational striped bass fishing using annual data from 1965 to 1979, measuring the effects
 8 of SAV levels on fish stocks, harvests, and social welfare. They estimated, for example, that a
 9 50% reduction in SAV from levels existing in the late 1970s (similar to current levels [CBP,
 10 2009]) would decrease the net social benefits from striped bass by roughly \$16 million (in 2007
 11 dollars).

12 In a separate analysis, Anderson (1989) developed an empirical dynamic simulation
 13 model of the effects of SAV changes on commercial blue crab harvests in the Virginia portion of
 14 the Chesapeake Bay. Applying the empirical model results, long-run (15-year) dynamic
 15 equilibria were estimated under baseline conditions (assuming SAV area constant at 1987 levels)
 16 and under conditions with “full restoration” of SAV (i.e., 284% increase). In equilibrium, the
 17 increase in annual producer surplus and consumer surplus with full restoration of SAV was
 18 estimated to be \$3.5 million and \$4.4 million (in 2007 dollars), respectively.

19 **Table 5.2-1.** Value of Commercial Landings for Selected Species in 2007
 20 (Chesapeake Bay Region)

State	Species	Value
Maryland	Blue crab	\$30,433,777
	Striped bass	\$5,306,728
	Clams or bivalves	\$5,007,952
	Sea scallop	\$2,808,984
	Oyster, Eastern	\$2,524,045
	Other	\$6,190,474
	Total	\$52,271,960

State	Species	Value
Virginia		
	Sea scallop	\$62,891,848
	Menhaden	\$25,350,740
	Blue crab	\$13,222,135
	Croaker, Atlantic	\$4,615,924
	Striped bass	\$3,834,906
	Clam, Northern Quahog	\$3,691,319
	Summer flounder	\$3,186,229
	Other	\$16,954,893
	Total	\$130,561,765

1 **Source:** NOAA (2007, August). “Annual Commercial Landings Statistics.”
2 (http://www.st.nmfs.noaa.gov/st1/commercial/landings/annual_landings.html)

3 One study examining the short-term effects of dissolved oxygen (DO) levels on crab
4 harvests is by Mistiaen et al. (2003). Focusing on three Chesapeake Bay tributaries—the
5 Patuxent, Chester, and Choptank rivers—this study estimated a “stress-availability” model
6 measuring the effects of DO levels on the availability of blue crabs for commercial harvest,
7 given the stock levels and number of fishing vessels. The model results indicated that, below a
8 threshold of 5 milligrams per liter (mg/L), reductions in DO cause a statistically significant
9 reduction in commercial harvest and revenues. For the Patuxent River alone, a simulated
10 reduction of DO from 5.6 to 4.0 mg/L was estimated to reduce crab harvests by 49% and reduce
11 total annual earnings in the fishery by \$275,000 (in 2007 dollars). However, this is an upper-
12 bound estimate because it does not account for changes in fishing effort that would likely occur,
13 and if the measured changes are due to migration of crab populations to other areas rather than to
14 crab mortality, then the broader net effects on crab harvests may also be considerably smaller.¹

15 In addition to affecting provisioning services through commercial fish harvests,
16 eutrophication in estuaries may also affect these services through its effects on the demand for
17 seafood. For example, a well-publicized toxic pfiesteria bloom in the Maryland Eastern Shore in
18 1997, which involved thousands of dead and lesioned fish, led to an estimated \$56 million (in

¹ The estimated relationship between harvest and DO is discontinuous at 5 mg/L. The size of the measured effect on harvests is relatively small below 5 mg/L and is zero above the 5 mg/L threshold; therefore, any sizable benefits would require DO to cross the 5 mg/L threshold. Moreover, the 5 mg/L threshold was an assumption of the model rather than a tested hypothesis, which raises additional questions about the accuracy of benefit estimates for changes across the threshold.

1 2007 dollars) in lost seafood sales for 360 seafood firms in Maryland in the months following the
2 outbreak (Lipton, 1999). Additional evidence regarding potential losses in provisioning services
3 due to eutrophication-related fish kills is provided by Whitehead et al. (2003) and Parsons et al.
4 (2006). The survey used in both studies was conducted with more than 5,000 respondents in
5 states bordering the Chesapeake Bay area and in North Carolina. The survey asked respondents
6 to consider how their consumption patterns would change in response to news about a large fish
7 kill caused by a toxic pfiesteria bloom. To address the fact that not all fish kills are the same, the
8 size and type of the described fish kill—either “major,” involving more than 300,000 dead fish
9 and 75% with pfiesteria lesions, or “minor,” involving 10,000 dead fish and 50% with
10 lesions—were randomized across respondents. Based on respondents’ stated behaviors, the
11 studies estimated reductions in consumer surplus per seafood meal ranging from \$2 to \$5.² The
12 survey also found that 42% of residents in the four-state area (i.e., Maryland, Virginia, Delaware,
13 and North Carolina) were seafood consumers and that the average number of seafood meals per
14 month among these consumers was between four and five. As a result, they estimated aggregate
15 consumer surplus losses of \$43 million to \$84 million (in 2007 dollars) in the month after a fish
16 kill.

17 **Cultural Services**

18 Estuaries in the eastern United States also provide an important and substantial variety of
19 cultural ecosystem services, including water-based recreational and aesthetic services. One of the
20 difficulties with quantifying recreational services from estuaries is that much of the national and
21 regional statistics are jointly collected and reported for estuarine and other coastal areas.
22 Nevertheless, even these combined statistics provide several useful indicators of recreational
23 service flows. For example, data from the Fishing, Hunting, and Wildlife-Associated Recreation
24 (FHWAR) indicate that, in 2006, 4.8% of the 16 and older population in coastal states from
25 North Carolina to Massachusetts participated in saltwater fishing (U.S. DOI, 2007). The total
26 number of days of saltwater fishing in these states was 26.1 million in 2006. Based on estimates
27 from Kaval and Loomis (2003), the average consumer surplus value for a fishing day was \$35.91
28 (in 2007 dollars) in the Northeast and \$87.23 in the Southeast. Therefore, the total recreational
29 consumer surplus value from these saltwater fishing days was approximately \$1.28 billion (in

² Surprisingly, these estimates were not sensitive to whether the fish kill was described as major or minor or to the different types of information included in the survey.

1 2007 dollars). Consumer surplus value is a commonly used and accepted measure of economic
2 benefit (see, for example, U.S. EPA, 2000b). It is the difference between (1) the maximum
3 amount individuals are, on average, willing and able to pay for a good, service, or activity (in
4 this case, a day of recreational fishing) and (2) the amount they actually pay (in out-of-pocket
5 and time costs). For recreation days, it is most commonly measured using recreation demand,
6 travel cost models.

7 Recreational participation estimates for several other coastal recreational activities are
8 also available for 1999 to 2000 from the National Survey on Recreation and the Environment.
9 Almost 6 million individuals age 16 and older participated in motorboating in coastal states from
10 North Carolina to Massachusetts, for a total of nearly 63 million days annually during 1999 to
11 2000. Using a national daily value estimate of \$32.69 (in 2007 dollars) for motorboating from
12 Kaval and Loomis (2003), the aggregate value of these coastal motorboating outings was \$2.08
13 billion per year. Almost 7 million individuals participated in birdwatching, for a total of nearly
14 175 million days per year, and more than 3 million individuals participated in visits to non-beach
15 coastal waterside areas, for a total of more than 35 million days per year. In contrast, less than 1
16 million individuals per year participated in canoeing, kayaking, or waterfowl hunting.

17 **Regulating Services**

18 Estuaries and marshes have the potential to support a wide range of regulating services,
19 including climate, biological, and water regulation; pollution detoxification; erosion prevention;
20 and protection against natural hazards (MEA, 2005c). It is more difficult, however, to identify
21 the specific regulating services that are significantly impacted by changes in nutrient loadings.
22 One potentially affected service is provided by SAV, which can help reduce wave energy levels
23 and thus protect shorelines against excessive erosion. Declines in SAV may, therefore, also
24 increase the risks of episodic flooding and associated damages to near-shore properties or public
25 infrastructure. In the extreme, these declines may even contribute to shoreline retreat, such that
26 land and structures are lost to the advancing waterline.

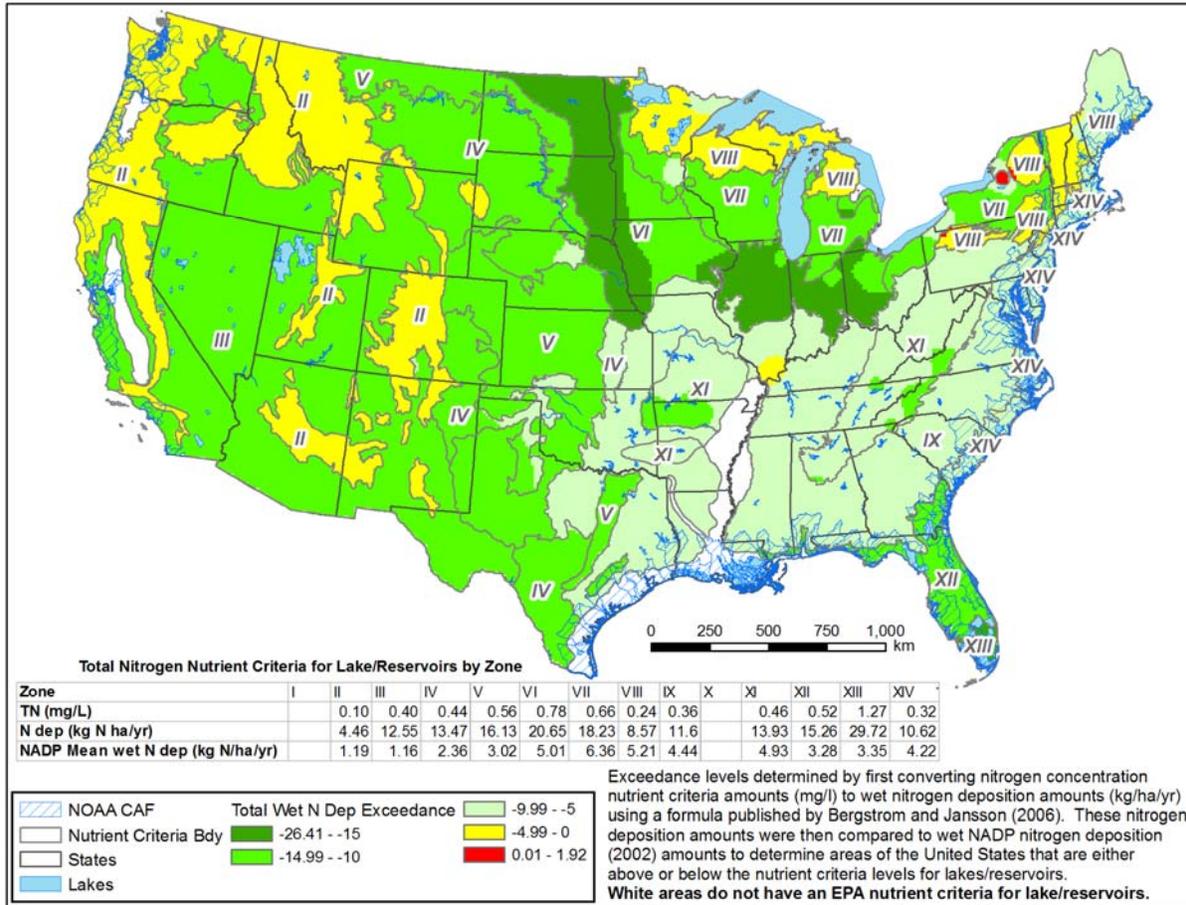
5.2.2 Characteristics of Sensitive Areas

It is the general consensus that nitrogen is the limiting element to primary production in coastal marine ecosystems in the temperate zone.

Howarth and Marino (2006) provide a comprehensive summary of the literature and scientific findings on eutrophication over the past 3 decades. That summary has led to the general consensus (1) that freshwater lakes and estuaries differ in terms of nutrient limitation as the cause of eutrophication, and (2) that nitrogen is the limiting element to primary production in coastal marine ecosystems in the temperate zone. The factors that make estuarine systems sensitive to nutrient enrichment are still weakly understood, but it is suggested that factors that influence the residence time of the estuarine waters and the complex interactions affecting nutrient and light limitation all play a role in defining sensitivity (Howarth and Marino, 2006). In general, ecosystems that are most vulnerable to nutrient enrichment from atmospheric nitrogen deposition are those that receive high levels of deposition relative to nonanthropogenic nitrogen loading, those that are nitrogen limited, or those that contain species that have evolved in nutrient-poor environments (U.S. EPA, 2008, Section 3.3)

The selection of case study areas specific to eutrophication began with national geographic information systems (GIS) mapping to identify sensitive areas. Spatial datasets were reviewed that included physical, chemical, and biological properties indicative of eutrophication potential in order to identify sensitive areas of the United States. Datasets included in the USGS National Water Quality Assessment (NAWQA) Program files, U.S. EPA STORage and RETrieval (STORET) database, NOAA Estuarine Drainage Areas data, and EPA's water quality standards nutrient criteria for rivers and lakes (see Appendix 6, Table 1.2-1). To define areas of national aquatic nutrient enrichment sensitivity, eutrophic estuaries from NOAA's Coastal Assessment Framework (CAF) and areas that exceed the nutrient criteria for lakes/reservoirs (U.S. EPA, 2002) were combined and compared to total nitrogen deposition. The resulting map revealed areas of highest potential sensitivity to nitrogen deposition as shown in **Figure 5.2-3**. These areas are identified in blue as nutrient-sensitive estuaries contained in NOAA's CAF, and in red in areas where deposition exceeds the nutrient criteria. Yellow areas indicate those areas that are below the nutrient criteria, but are within 5 kilograms (kg) N/ha/yr of exceeding it. While this map delineates those regions that are sensitive to deposition by virtue of bedrock and

1 topography, it may not represent regions with perched waterbodies that receive nitrogen
 2 deposition.



3
 4 **Figure 5.2-3.** Areas potentially sensitive to aquatic nutrient enrichment.
 5 **Note:** The estimated atmospheric deposition loads in these areas are based solely on wet
 6 deposition, not dry deposition data. Blue areas are nutrient-sensitive estuaries contained in
 7 NOAA’s Coastal Assessment framework. Areas in red represent locations where deposition
 8 exceeds the nutrient criteria. Yellow areas indicate areas that are below the nutrient criteria, but
 9 are within 5 kg N/ha/yr of exceeding the criteria. While this map delineates those regions that are
 10 sensitive to deposition by virtue of bedrock and topography, it may not represent regions that
 11 have perched waterbodies that receive nitrogen deposition.

5.2.3 Case Study Selection

12
 13 Recommended case study areas are presented in the ISA (U.S. EPA, 2008, Sections 3.2,
 14 3.3, 3.4, 4.2, 4.3, 4.4, Annex B, and Annex C) as candidates for risk and exposure assessments.
 15 The Ecological Effects Subcommittee of the Advisory Council on Clean Air Compliance
 16 Analysis also made recommendations (see Appendix 6, Table 1.2-3). These recommendations, in

1 tandem with the areas identified in the national characterization previously described, were used
2 to select case study areas for this Risk and Exposure Assessment.

3 Two regions were selected for case study analysis to which a common methodology
4 could be applied—the Chesapeake Bay and the Pamlico Sound. Both estuaries were selected
5 primarily based on the availability of research data. For aquatic nutrient enrichment, special
6 emphasis was given to the Chesapeake Bay region because it has been the focus of many
7 previous studies and modeling efforts, and it is currently one of the few systems within the
8 United States in which economic-related ecosystem services studies have been conducted. The
9 Pamlico Sound, an economically important estuary due to its fisheries, has been studied and
10 modeled greatly by the local universities and has also been known to exhibit symptoms of
11 extreme eutrophication. Factors including availability of atmospheric deposition data, existing
12 water quality modeling, and generalization opportunities for risk analysis from results were
13 considered in choosing these case study areas. Other candidate estuarine systems could be
14 evaluated for potential future analyses, while freshwater ecosystems in the western United States
15 would most likely require a separate analysis. Because the Chesapeake Bay and Pamlico Sound
16 are fed by multiple river systems, the case study was scaled to one main stem river for each
17 system: the Potomac River/Potomac Estuary and the Neuse River/Neuse River Estuary. Details
18 on each basin are provided in Appendix 6, Sections 1.2.3 and 1.2.4, respectively.

19 The Potomac River contains diverse watersheds in terms of topography, elevation (e.g.,
20 extending into the Shenandoah Mountains), and nutrient point and nonpoint sources (e.g.,
21 forestland, farmland, and the Washington, DC, metropolitan area). The 14,670 mi² (38,000
22 kilometers [km]²) basin lies in five geological provinces: the Appalachian Plateau, Ridge and
23 Valley, Blue Ridge, Piedmont Plateau, and Coastal Plain. The watershed is approximately 12%
24 urbanized, 36% agricultural use, and 52% forested. Atmospheric deposition has been reported to
25 contribute from 5% to 15%–20% of the basin’s total nitrogen load (U.S. EPA, 2000; Boyer et al.,
26 2002).

27 The Neuse River is the longest river in North Carolina and is a mainstem river to the
28 Pamlico Sound—one of the two largest estuaries on the Atlantic Coast. The drainage area for the
29 basin is approximately 14,210 mi² (36,804 km²) (NC DENR, 2002). The Neuse River watershed
30 has a population of approximately 1,320,379, according to the 2000 census. Fifty-six percent of
31 the land in the basin is forested, and approximately 23% is in cultivated cropland. There are

1 134,540 estuarine hectares (332,457 acres) classified for shellfish harvesting (Class SA
2 [shellfishing]) in the Neuse River Estuary. Atmospheric deposition is believed to play a role in
3 nutrient loading to the Neuse River and Pamlico Sound. According to Whitall and Paerl (2001),
4 atmospheric deposition accounts for approximately 24% of the Neuse River watershed's total
5 nitrogen loading. Of these atmospheric deposition measurements, it is expected that the
6 contributions will be greater from reduced forms of nitrogen than from oxidized forms because
7 of the large amounts of agriculture within the watershed. One of the reasons for selecting the
8 Neuse River/Neuse River Estuary Case Study Area is to evaluate the impact of a NO_x-based
9 standard on an area dominated by reduced forms of nitrogen.

10 **5.2.4 Current Conditions in the Case Study Areas**

11 The Chesapeake Bay is the largest estuary in the United States and has a complex
12 ecosystem of important habitats and food webs. The Potomac River is the second largest of five
13 major rivers that feed the Chesapeake Bay. Most of the Chesapeake Bay's waters are degraded.
14 Remediation goals over multiple categories were set forth in the Chesapeake 2000 Commitment,
15 an agreement between the heads of several state and commission stakeholders (Chesapeake Bay
16 Executive Council, 2000). Because there are numerous indices and categories in which
17 remediation goals have been set, the reader is instructed to view the Chesapeake Bay Program's
18 Remediation Web site for specific inquiries:
19 <http://www.chesapeakebay.net/bayrestoration.aspx?menuitem=13989>. In 2007, it was 21% of the
20 way toward meeting water quality goals (e.g., 40% reduction in nitrogen and phosphorus over
21 1987 levels). The Chesapeake Bay's current habitats and lower food web are at 44% of desired
22 levels (e.g., increased number of oysters, restored area of wetlands). Many of the Chesapeake
23 Bay's fish and shellfish populations are below historic levels. Currently, the Chesapeake Bay's
24 fish and shellfish are at 52% of desired levels (e.g., counts of blue crabs, oysters, striped bass).
25 Runoff from winter and spring rains delivers loads of sediment and nutrient pollutants that drive
26 summer water quality conditions. Past observations reveal that summer weather conditions also
27 contribute to summer water quality when intense storms increase erosion. Nutrients reach the
28 Chesapeake Bay from point and nonpoint source discharges and atmospheric deposition from a
29 570,000-mi² airshed (CBP, 2009) The National Water Quality Assessment states that although
30 nitrogen and phosphorus occur naturally, elevated concentrations of nutrients often result from

1 human activities. Atmospheric deposition from combustion of fossil fuels alone accounts for
2 32% of nitrogen inputs (<http://pubs.usgs.gov/circ/circ1166/circ1166.pdf>). Although NAWQA
3 states that the water quality concentration of nutrients in the Potomac River watershed does not
4 pose a direct exposure threat to human health or wildlife, excessive nitrogen or phosphorus in
5 streams can cause eutrophication. It is the condition of the Potomac Estuary (as a component of
6 the Chesapeake Bay) and its eutrophication potential that are the focus of the Aquatic Nutrient
7 Enrichment Case Study.

8 Eutrophication became a water quality concern in the lower Neuse River watershed in the
9 late 1970s and early 1980s, and fish kills, algal blooms, and correspondingly high levels of
10 chlorophyll *a* prompted the State of North Carolina to place the Neuse River Estuary on the
11 1994, 1996, 1998, and 2000 303(d) List of Impaired Waters.

12 To assess current conditions for the Potomac River/Potomac Estuary Case Study Area
13 and Neuse River/Neuse River Estuary Case Study Area, it was necessary to have atmospheric
14 deposition data available to input to SPARROW. The deposition data used for the Aquatic
15 Nutrient Enrichment Case Study are based on the 2002 CMAQ model year and NADP
16 monitoring data; therefore, current conditions for this case study evaluated ecosystem responses
17 for the year 2002. In both the Potomac River/Potomac Estuary Case Study Area and the Neuse
18 River/Neuse River Estuary Case Study Area, the best attempts were made to use monitoring and
19 modeling data from that time period (2002). Annual averages for 2002 were used in this study.

20 ***5.2.4.1 Potomac River and Potomac Estuary Current Conditions***

21 **SPARROW Assessment.** For the current condition 2002 analysis of the Potomac
22 River/Potomac Estuary Case Study Area, an estimated 40,770,000 kg of total nitrogen was
23 deposited in the Potomac River watershed for an average deposition of 12.9 kg N/ha/yr. **Figure**
24 **5.2-4** through **Figure 5.2-6** reveal highly different spatial patterns in oxidized, reduced, and total
25 nitrogen atmospheric deposition across the watershed. Note that the scales across the three
26 figures use the same increments and colors, so that they can be compared directly.

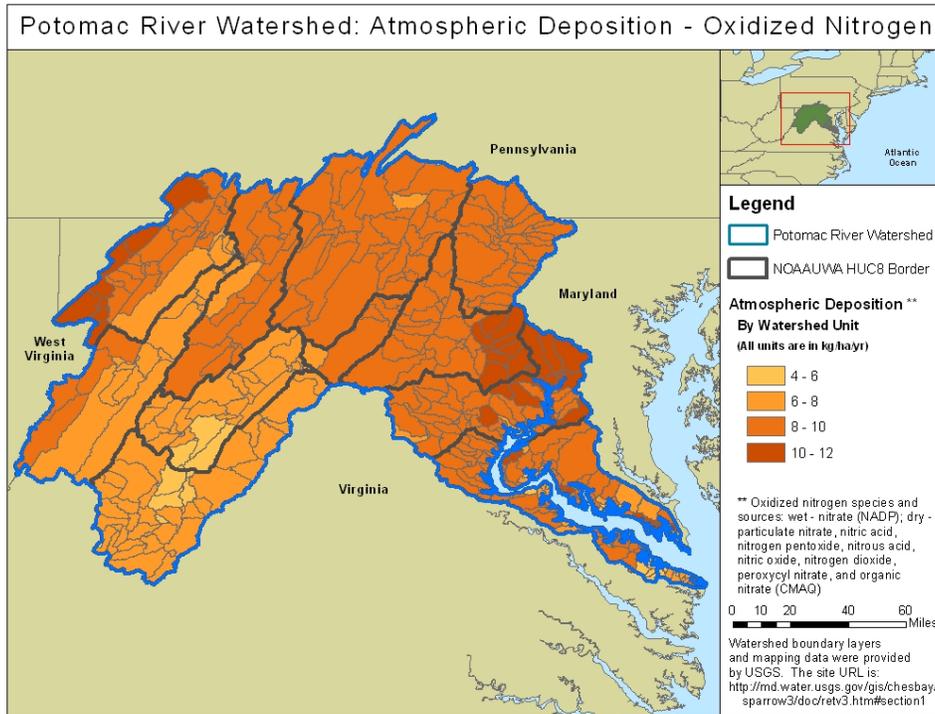
27 Application of a previously calibrated version of the SPARROW model for the
28 Chesapeake Bay watershed provides estimates of the incremental yield derived within each
29 catchment of the Potomac River watershed, as well as estimates of the delivered yield (i.e., the
30 fraction of the incremental flux that ultimately reaches the estuary) (**Figure 5.2-7**). (Details on

1 the use of the Version 3 Chesapeake Bay SPARROW model can be found in Appendix 6.) By
2 looking at catchment-scale results, the spatial variability among the loading contributions across
3 the watershed can be shown. Differences between the incremental and delivered yields reflect the
4 instream losses that occur as the load from each catchment travels downstream to the target
5 estuary.

6 For this application and analysis of the 2002 current condition case, SPARROW was
7 used to model the loads from the Potomac River and its watershed to the upper portions of the
8 Potomac Estuary. The most downstream modeled catchment in the analysis lies downstream of
9 several major point sources between Washington, DC, and the mixing zone of the estuary. These
10 point sources were major contributors of nutrients to the estuary, and by including them in the
11 analysis, a more accurate load from the Potomac River watershed is defined rather than if the
12 modeling stopped at the fall line of the river. Direct runoff from catchments surrounding the
13 estuary and direct deposition to the estuary were not considered in this preliminary model
14 application. The majority of the nitrogen loading to the estuary was expected to derive within the
15 Potomac River watershed because of overall larger land area and applications of fertilizer and
16 manure. Additionally, the major point sources to the Potomac Estuary were included in the most
17 downstream watersheds at the mouth of the estuary modeled in this application.

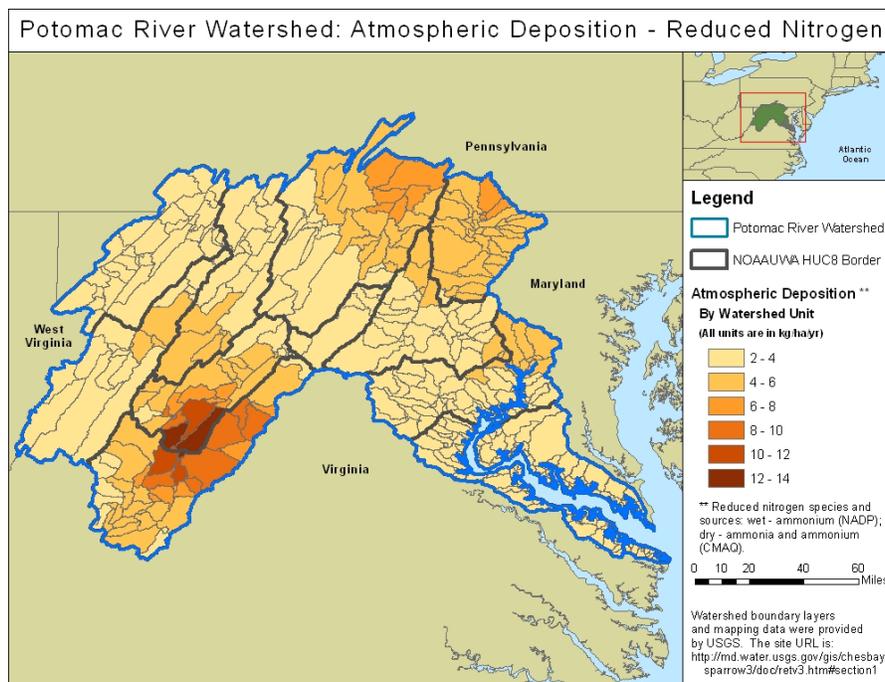
18 Overall, the SPARROW model produced an estimate of total nitrogen loading to the
19 Potomac Estuary of 36,660,000 kg N/yr. The atmospheric deposition load was estimated at
21 7,380,000 kg N/yr to the estuary, or 20% of the total
23 loading. These modeling estimates are consistent with
25 previous modeling estimates for the system (Preston and
27 Brakebill, 1999). The TN_s resulting from this loading was
29 approximately 3.4 mg/L.

SPARROW modeling for 2002 predicts that atmospheric deposition was 20% of the total nitrogen loading to the Potomac River's estuary, producing an TN_s of 3.4 mg/L.



1
2
3

Figure 5.2-4. Atmospheric deposition yields of oxidized nitrogen over the Potomac River and Potomac Estuary watershed.



4
5
6

Figure 5.2-5. Atmospheric deposition yields of reduced nitrogen over the Potomac River and Potomac Estuary watershed.

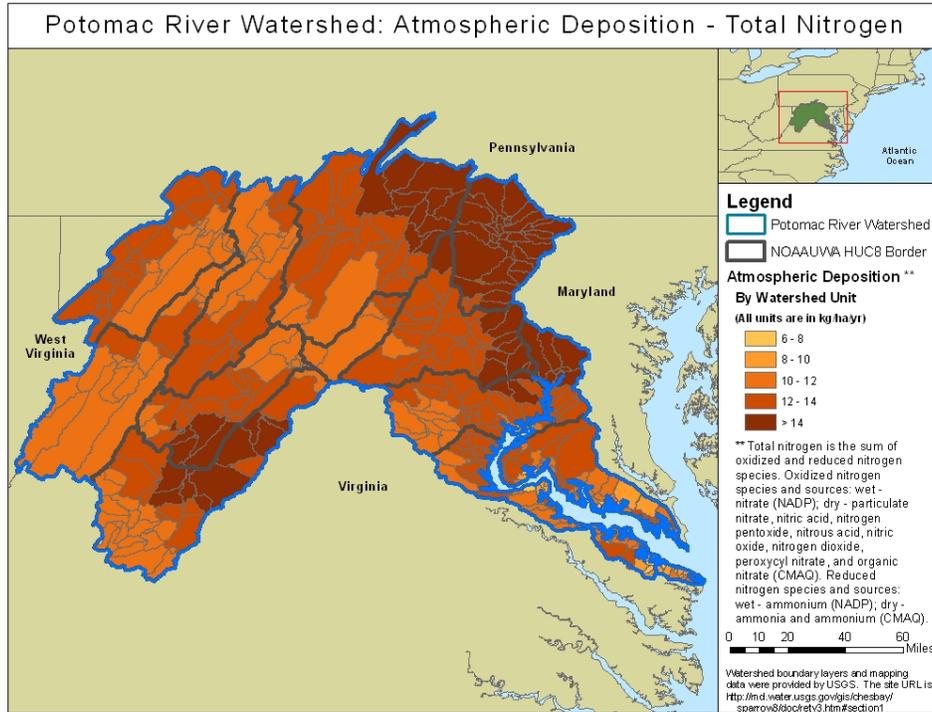


Figure 5.2-6. Atmospheric deposition yields of total nitrogen over the Potomac River and Potomac Estuary watershed.

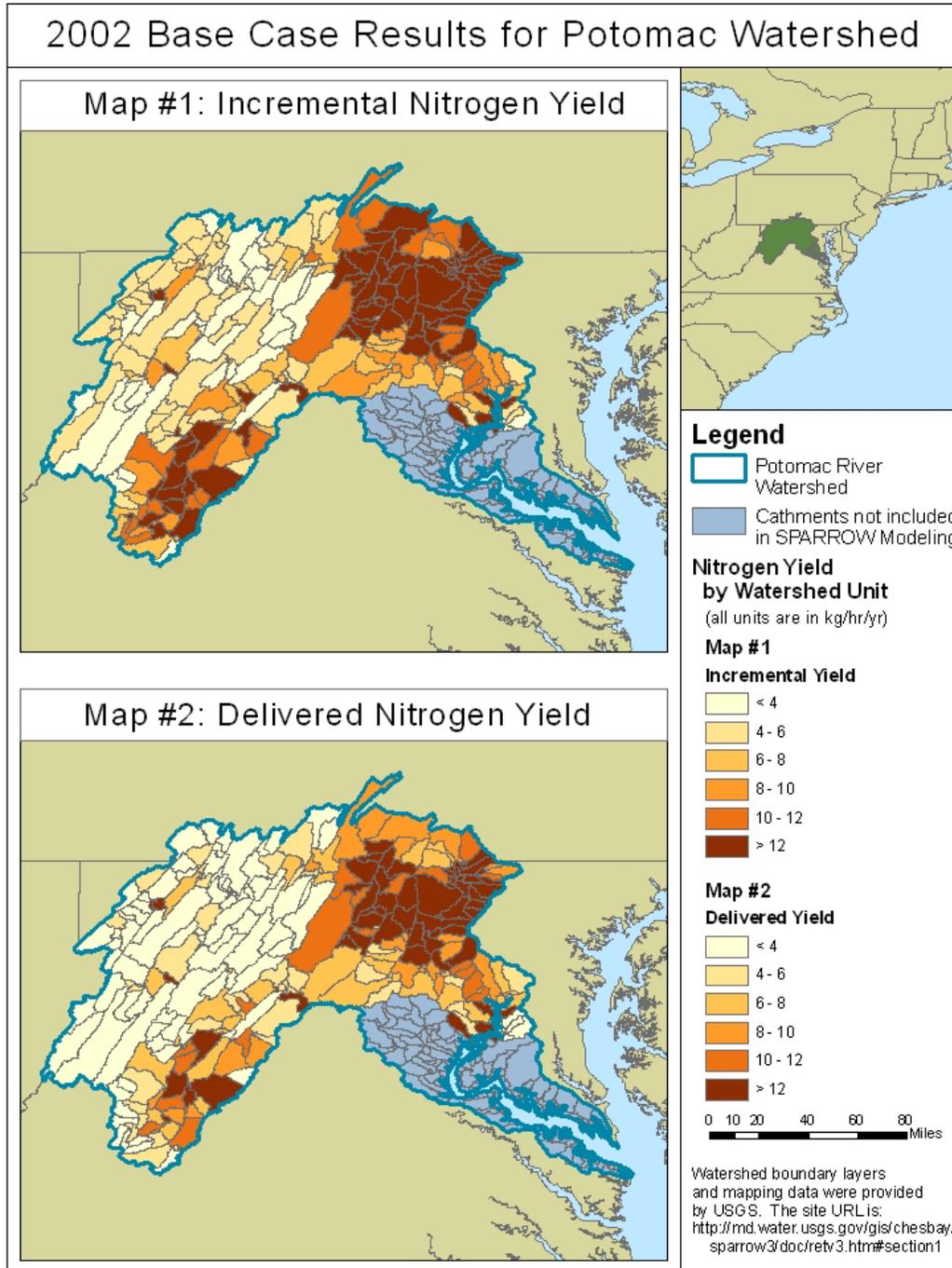
ASSETS EI Assessment. An ASSETS EI was completed for the Potomac Estuary in a 2006 NOAA project on the Gulf of Maine (Bricker et al., 2006) using 2002 data to determine the scoring. That assessment showed that the system has a high susceptibility to pressures and a high score for nutrient inputs, resulting in a *High* OHI score. Individual scores for the primary and secondary indicators varied but resulted in an overall *High* OEC score. The score of *Improve Low* for the DFO is based on the expectations that future nutrient pressures will decrease and there will be significant population and development increases.

For the Aquatic Nutrient Enrichment Case Study, the ratings for the nutrient inputs and OEC were re-created and verified using methods consistent with the 2007 NEEA

The ASSETS EI for the 2002 Potomac River estuary current condition scenario is *Bad*.

Update, which included separate area-weighted consideration of the tidal fresh, mixing, and saltwater zones within the estuary (Bricker et al., 2007). Index scores for the updated analysis were compiled using the scoring methods and matrices as shown in Figures 2.2-6 and 2.2-7 of Appendix 6. Combination of the primary and secondary scores (both *High*) provided an overall OEC score of *High*, which agreed with the original analysis. The OHI score (confirmed with the

1 modeled nitrogen load from the 2002 SPARROW application) and the DFO scores remain the
 2 same as in the original analysis. Therefore, the ASSETS EI for the 2002 current condition
 3 scenario is *Bad*.

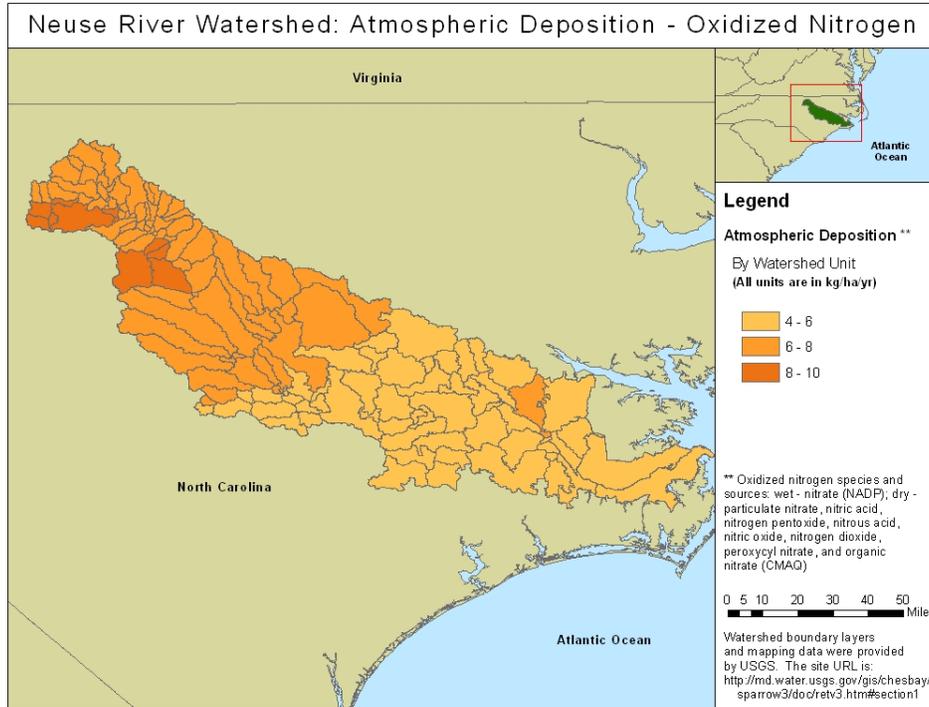


4
 5 **Figure 5.2-7.** Total nitrogen yields from all sources as predicted using version 3 of the
 6 Chesapeake Bay SPARROW application with updated 2002 atmospheric deposition inputs.

1 **5.2.4.2 Neuse River and Neuse River Estuary Current Conditions**

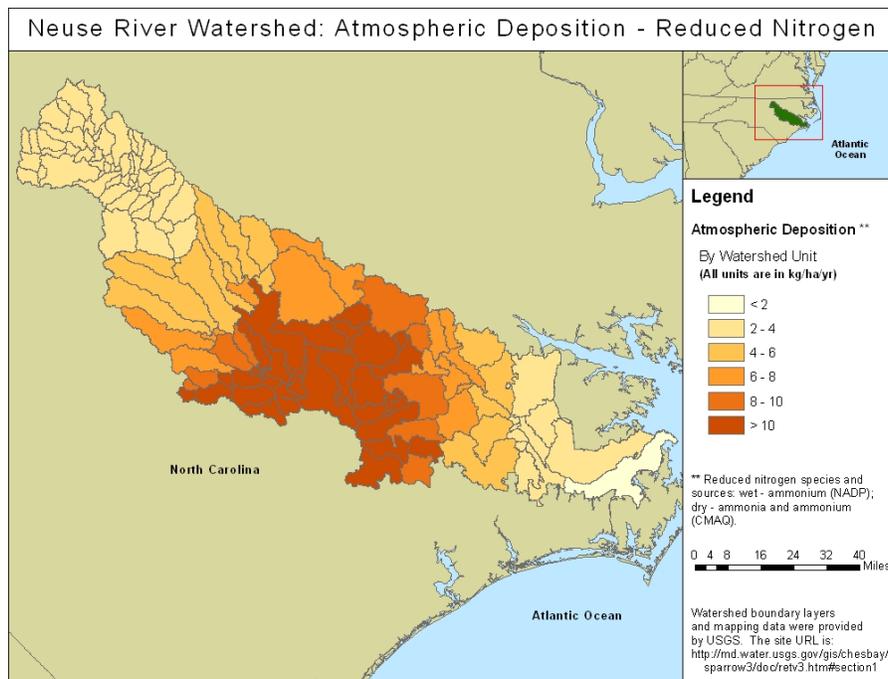
2 The current condition 2002 analysis of the Neuse River and Neuse River Estuary used
3 recently released data from the USGS to calibrate a new SPARROW application for 2002 to the
4 Neuse watershed. (Because of a limited number of calibration points within the Neuse watershed
5 itself, the SPARROW model assessment was expanded to include the Tar-Pamlico and Cape
6 Fear River basins, providing a total of 41 calibration points on which to base the SPARROW
7 model.) Developing the ASSETS EI for the Neuse River Estuary proved to be a greater challenge
8 than for the Potomac Estuary due to data sources being less consolidated and more varied.

9 **SPARROW Assessment.** **Figure 5.2-8** through **Figure 5.2-10** present the atmospheric
10 deposition inputs used within the modeling effort. For 2002, an estimated 18,340,000 kg of total
11 nitrogen was deposited in the Neuse River watershed for an average deposition of 14.0 kg
12 N/ha/yr. The model was based on total nitrogen loads from deposition, but oxidized and total
13 reactive nitrogen yields are also presented to highlight source information within the watershed.
14 The Neuse River watershed is the location of major agricultural operations focusing on swine
15 facilities. These operations are evident in the high levels of reduced nitrogen found within the
16 south-central catchments of the watershed (**Figure 5.2-9**).



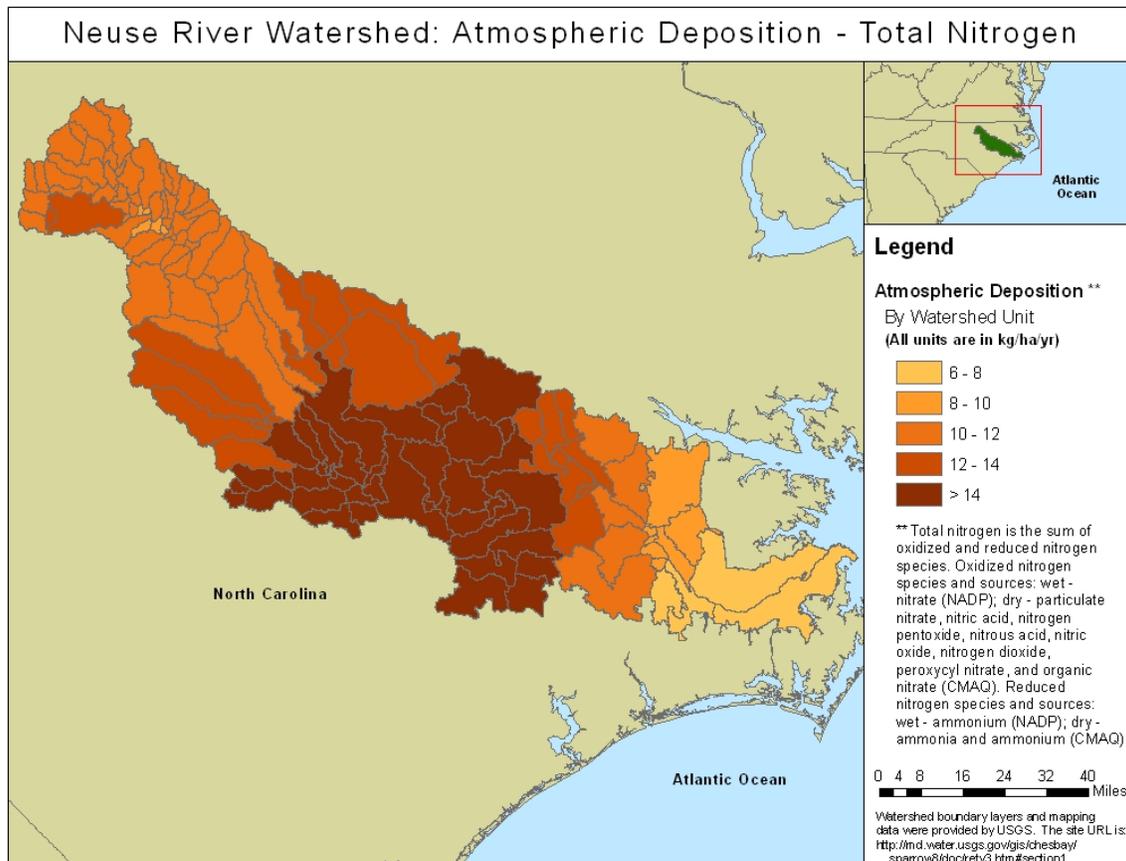
1
2
3

Figure 5.2-8. Atmospheric deposition yields of oxidized nitrogen over the Neuse River and Neuse River Estuary watershed.



4
5
6

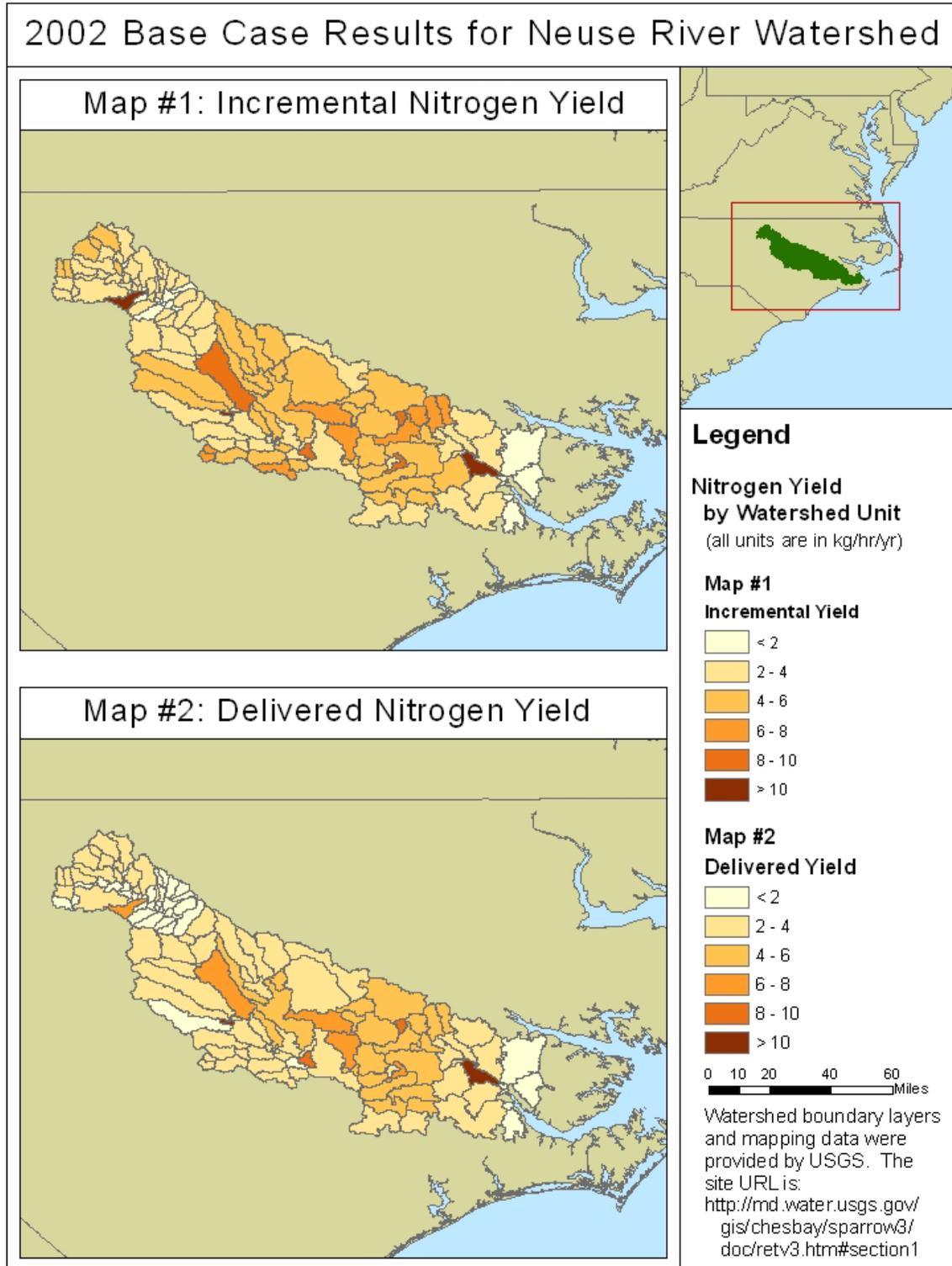
Figure 5.2-9. Atmospheric deposition yields of reduced nitrogen over the Neuse River and Neuse River Estuary watershed.



1
2 **Figure 5.2-10.** Atmospheric deposition yields of total nitrogen over the Neuse River and
3 Neuse River Estuary watershed.

4 As with the Potomac River and Potomac Estuary watershed results, the Neuse River and
5 Neuse River Estuary SPARROW application modeled watershed loads to the upper edges of the
6 estuary. Both the incremental and delivered yields are presented in **Figure 5.2-11**. The total
7 nitrogen load estimated to enter the estuary from the Neuse River is 4,380,000 kg N/yr, equating
8 to a TN_s of 1.11 mg/L. Atmospheric deposition was estimated
9 to contribute 1,150,000 kg N/yr, or 26% of the total load.
10
11 These estimates fall in line with instream monitoring data and
12 previous loadings from the Neuse River estimated at 9.61
13 million pounds or 4,359,000 kg N/yr (Spruill et al., 2004).
14
15
16
17
18

SPARROW modeling for 2002 predicts that atmospheric deposition was 26% of the total nitrogen loading to the Neuse River's estuary, producing a TN_s of 1.1 mg/L.



1
2
3

Figure 5.2-11. Total nitrogen yields from all sources predicted by a SPARROW application for the Neuse, Tar-Pamlico, and Cape Fear watersheds with 2002 data inputs.

2 **ASSETS EI Assessment.** Previous work was completed by NOAA using the ASSETS
 4 EI on the Neuse River Estuary as part of the NEEA
 6 Update (Bricker et al., 2007). The exact source of the
 8 load estimate and the exact timeframe of the data used
 9 to calculate the ASSETS EI are still unknown at this time, although the data should fall between
 10 2000 to 2002 (S. Bricker, personal communication, 2008). That analysis revealed a
 11 *Highly/Moderately Influenced* or *High* score for influencing factors where the nitrogen load was
 12 ranked as *Moderate to High*, resulting in a *Bad* overall ASSETS score for the estuary.

Combining the OEC, OHI, and DFO indices results in an overall ASSETS EI for the Neuse River Estuary for 2002 of <i>Bad</i> .
--

13 To develop an updated ASSETS EI specific to the 2002 baseline for this assessment,
 14 available data from multiple sources, including the Neuse River Estuary Modeling and
 15 Monitoring Project, were combined to form a 2002 OEC score. Because both the chlorophyll *a*
 16 and harmful algal bloom data were available and overwhelmingly pointed to a system with both
 17 *High* primary and secondary scores, a *High* OEC rating is given with confidence for 2002. The
 18 *High* susceptibility ranking, combined with the total nitrogen loads estimated by the SPARROW
 19 assessment, rank the OHI as *High* as well. The DFO set during the 2007 NEEA Update remains
 20 unchanged, with a ranking of *Worsen High* due to nutrient reductions from improved
 21 management practices in recent years being offset by increases in human populations and factors
 22 related to swine production (Burkholder et al., 2006). Combining the three indexes results in an
 23 overall ASSETS EI for the Neuse River/Neuse River Estuary Case Study Area for 2002 of *Bad*.

24 **5.2.5 Degree of Extrapolation to Larger Assessment Areas**

25 Selection of the analysis method for aquatic nutrient enrichment considered applications
 26 beyond a small number of case studies. The chosen method, consisting of a combination of
 27 SPARROW modeling for nitrogen loads and an assessment of estuary conditions under the
 28 NOAA ASSETS EI, provides a highly scalable and widely applicable analysis method. Both
 29 components have been applied on a national scale—the national nutrient assessment using
 30 SPARROW (Smith and Alexander, 2000) and the NEEA using the ASSETS EI (Bricker et al.,
 31 1999, 2007). Additionally, both have been used on a smaller scale. These previous analyses
 32 supply a large body of work—data, methods, and supporting experts—to draw from when
 33 conducting additional analyses or updating past applications.

1 Requirements for applying this method to other systems include mandatory data inputs,
2 the ability to formulate a SPARROW application on a reliable stream network, and an estuary
3 under suspicion of eutrophication. Data requirements and model formulations have been
4 described and detailed throughout this report.

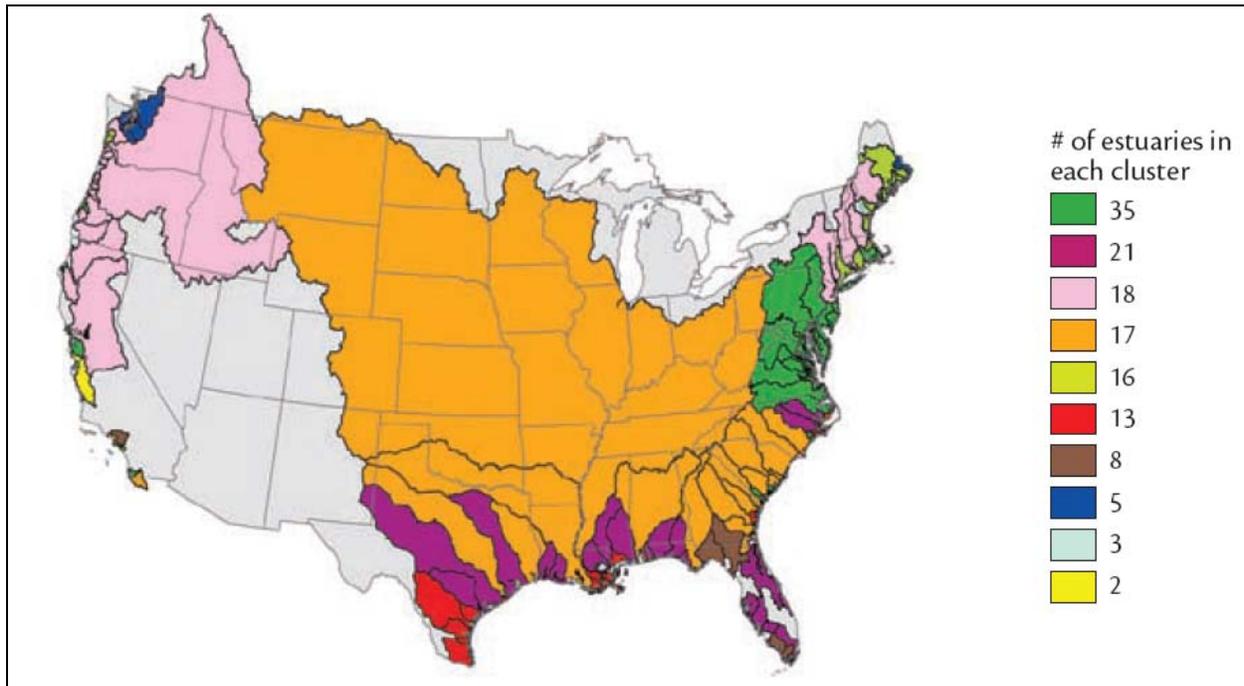
5 The method is not currently designed to assess eutrophication impacts on inland waters;
6 however, a separate analysis was conducted of the extent inland waters exceed national nutrient
7 criteria for nitrogen. Results are presented in the GIS analysis for sensitive areas of the United
8 States that are identified in Appendix 6. SPARROW modeling can be applied to determine
9 nitrogen loadings to an inland waterway, but the ASSETS EI would not apply, and as such, the
10 indicators and overall likelihood of eutrophication could not be assessed. For these inland waters,
11 an alternate methodology would be necessary to examine the effects of changing nitrogen loads
12 within the waterbody. A variety of methods could possibly be applied, including empirical
13 relationships or dynamic modeling. An additional case study, the Aquatic Acidification Case
14 Study, examines the effects of aquatic acidification on inland waters using dynamic modeling.

15 The scalability of the methods and approaches taken in these case studies will rely on the
16 ability to group estuaries across the country into patterns of similar behavior either in terms of
17 nitrogen sources or eutrophication effects. In 2003 and 2004, NOAA and the Kansas Geological
18 Survey conducted a series of workshops to develop a type classification system for the 138
19 estuarine systems assessed in the original NEEA (Bricker et al., 1999). Participants considered
20 70 classification variables for grouping the estuarine systems. These variables included 51
21 physical characteristics (e.g., estuary depth and volume, tidal range, salinity, nitrogen and
22 phosphorus concentrations, estimates of flushing time, evaporation), 10 perturbation
23 characteristics (e.g., population in watershed, estimates of nutrient loading), and nine response
24 characteristics (e.g., SAV loss, presence of nuisance, toxic blooms). Ultimately, the workgroup
25 selected five variables (i.e., depth, openness of estuary mouth, tidal range, mean annual air
26 temperature, the log of freshwater inflow/estuarine area) deemed to be the most critical physical
27 and hydrological characteristics influencing nutrient processing and the expression of eutrophic
28 symptoms in a waterbody. Based on these five variables, the 138 estuarine systems were
29 classified into 10 groups (**Table 5.2-2; Figure 5.2-12**). The two estuary systems included in this
30 case study, Potomac River Estuary and Neuse River Estuary systems, were in groups one and
31 nine, respectively (Bricker et al., In prep).

1 **Table 5.2-2.** Typology Group Categorizations

Group	Number of Systems	Overriding Characteristics
Group 0	13	Low freshwater inflow:estuarine area ratio; low depth; low estuary mouth openness
Group 1	35	Medium depth; medium estuary mouth openness; high annual air temperature
Group 2	5	High depth; low annual air temperature
Group 3	8	High estuary mouth openness; high depth
Group 4	18	Low estuary mouth openness; high freshwater inflow:estuarine area ratio; low annual air temperature
Group 5	3	High estuary mouth openness; high depth
Group 6	2	High depth; high estuary mouth openness
Group 7	16	High tidal range; medium estuary mouth openness; low annual air temperature
Group 8	17	High freshwater inflow:estuarine area ratio; low depth
Group 9	21	Low depth; medium estuary mouth openness; high annual air temperature

2



3

4 **Figure 5.2-12.** Preliminary classifications of estuary typology across the nation
 5 (Bricker et al., 2007).

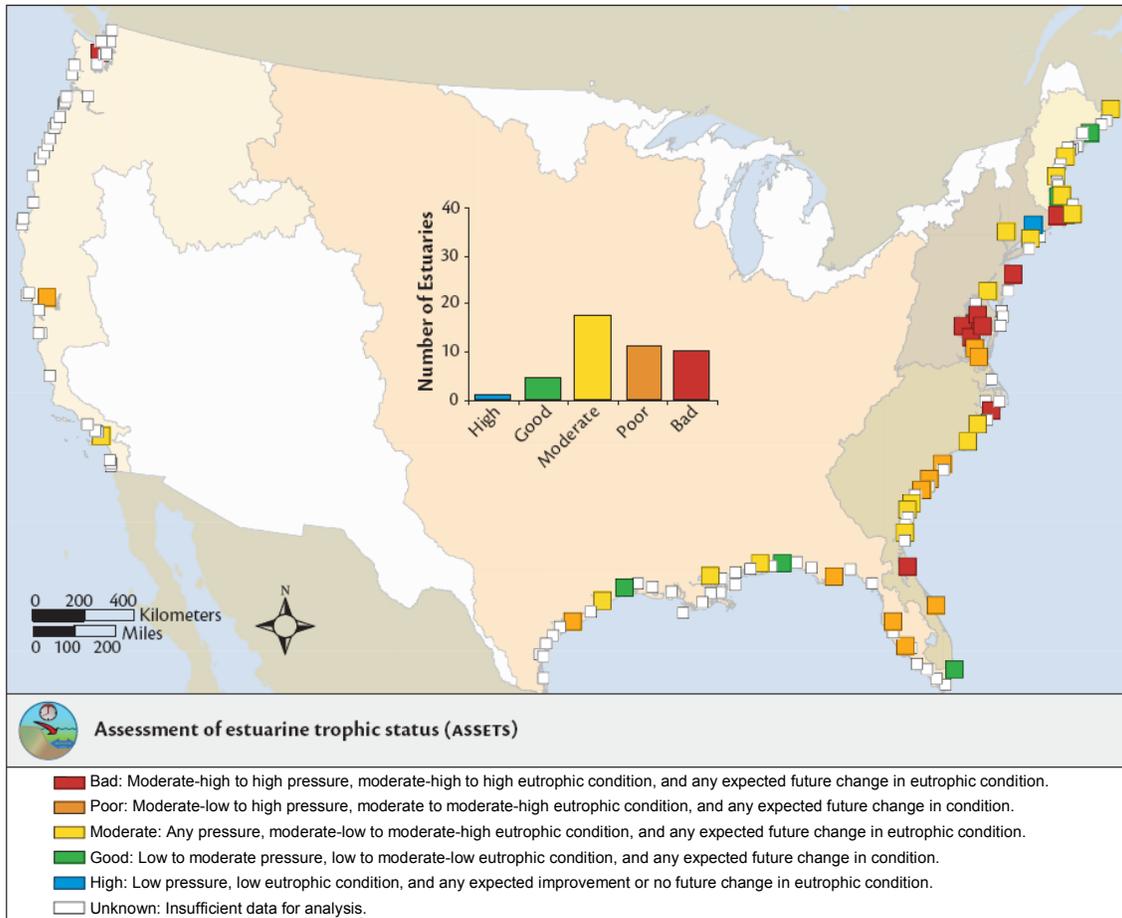
6 Given that the response curve of the OEC to total nitrogen (TN) is expected to change
 7 shapes with different values of susceptibility, the typology classes thus defined in **Table 5.2-2**

1 provide an opportunity to assess the validity of this expectation. The first step in assessing this
2 statement would be to examine the nutrient loadings in other estuaries that fall within groups 1 or
3 9, the groups corresponding to the two case studies. Once the shape and behavior of the response
4 curve for the estuary grouping is confirmed, work can begin to scale the results between estuaries
5 of that group. The ASSETS EI rating of an estuary may also be considered within this analysis.

6 Scaling of results will also need to account for the response of the watershed to
7 atmospheric nitrogen deposition inputs. If SPARROW continues to be used, either through the
8 in-development Web-enabled national SPARROW application or through regional or site-
9 specific applications, the shape of the response curve will be determined by the model and its
10 parameters. If a different approach is taken to developing total nitrogen loadings, then the
11 systems will need to be grouped according to the shape and behavior of the response curve.
12 Additional consideration should be given to the magnitude of the percentage contributions of the
13 atmospheric deposition to the total nitrogen load to the watershed and the resulting total nitrogen
14 load to the estuary.

15 **5.2.6 Current Conditions for Other/Additional Estuaries**

16 For 48 systems for which an ASSETS EI rating was developed in the 2007 NEEA
17 Update, only one system was rated as *High* (i.e., Connecticut River), while five were rated as
18 *Good* (i.e., Biscayne Bay, Pensacola Bay, Blue Hill Bay, Sabine Lake, Boston Harbor). Eighteen
19 were rated as *Moderate*, and 24 systems were rated as *Poor* or *Bad* (**Figure 5.2-13**). Comparing
20 the spatial distribution of these results to the preliminary typology groups described in the
21 previous section, the majority of estuaries in Group one, the group to which the Potomac Estuary
22 belongs, received scores of *Bad*. These conditions provide an opportunity to extrapolate between
23 the analysis methods and results determined for the Potomac Estuary and the other estuaries
24 matching in typology and current condition. For Group nine, to which the Neuse River Estuary
25 belongs, a greater range in ASSETS EI scores is found. Extrapolation of results within this group
26 must be examined in greater detail.



1
2 **Figure 5.2-13.** ASSETS EI scores for 48 systems examined in the 2007 NEEA Update
3 (Bricker et al., 2007).

4 **5.2.7 Ecological Effect Function for Aquatic Nutrient Enrichment**

5 Alternative effects levels were assessed for both the Potomac River and Neuse River
6 watersheds separately by applying percentage reductions to the oxidized nitrogen loads in the
7 estimated atmospheric deposition. Model estimates then relied on the SPARROW models used
8 (for the Potomac River) or developed (for the Neuse River) for the 2002 current condition
9 analysis to determine how the changing atmospheric inputs (i.e., total nitrogen load evaluated
10 with changes in oxidized nitrogen deposition, NO_x) affect the overall total nitrogen load to the
11 estuary of interest. These results were used to create the response curve relating instream total
12 nitrogen concentrations to atmospheric deposition loads as first described in Appendix 6, Section
13 2.2.3. The second response curve described in Section 2.2.3 was defined for the alternative
14 effects level analysis using historical data compilations of OEC scores and instream total
15 nitrogen concentrations while holding the susceptibility portion of the OHI (at its 2002 current

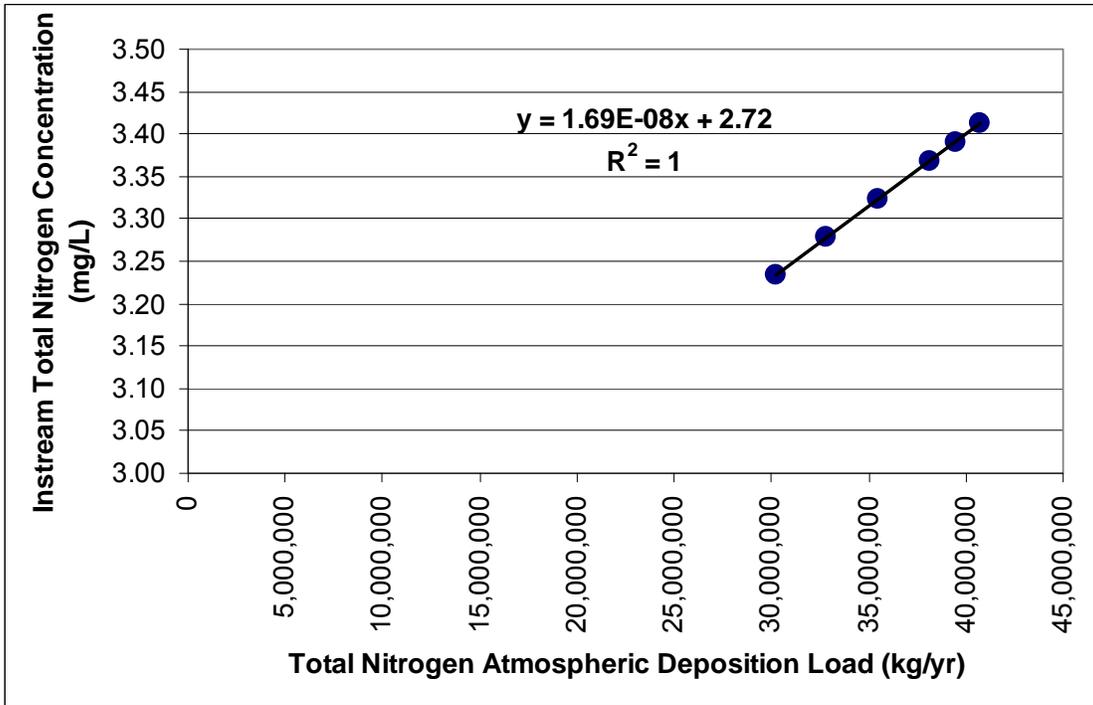
1 condition level— in both cases a ranking of *High*) and the DFO constant (at a ranking of *No*
2 *Change* [3]).

3 Upon creation of the two response curves, the back calculation coded program described
4 in Appendix 6, Section 2.2.3 (referred to as *BackCalculation* through the remainder of this
5 document) was applied to the curves with the intent of defining the atmospheric loads that are
6 needed to improve the ASSETS EI from a score of *Bad* (1) to *Poor* (2), *Moderate* (3), *Good* (4),
7 or *High* (5). These improvements represent improvements by 1, 2, 3, and 4 categories.

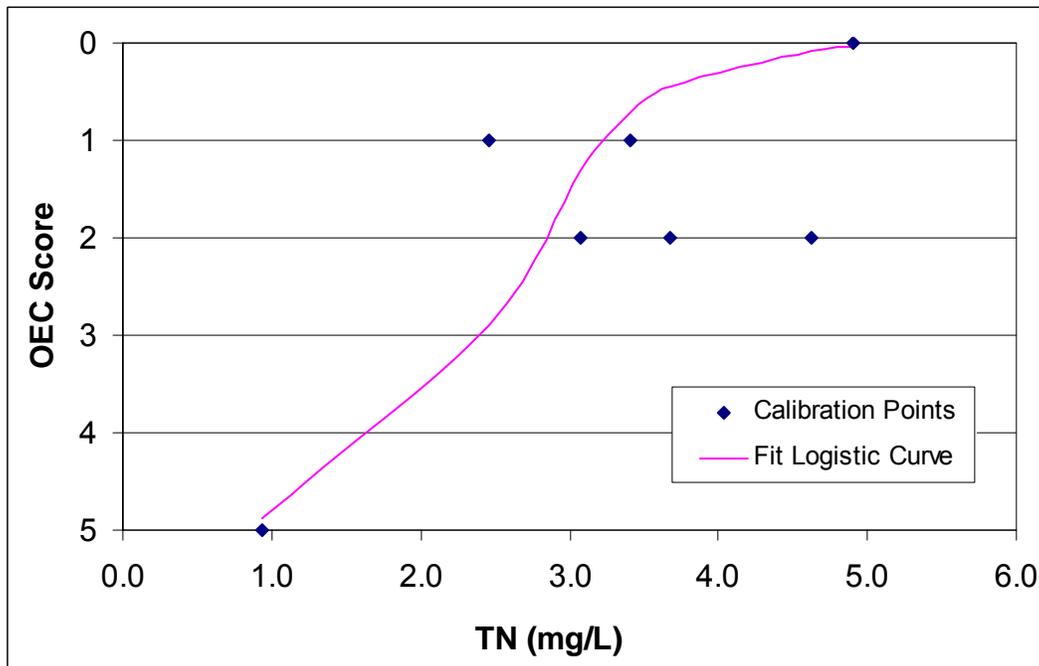
8 5.2.7.1 *Potomac River and Potomac Estuary*

9 Beginning with the data and model used for the current condition analysis, the
10 atmospheric deposition inputs derived from national coverage of CMAQ and NADP data were
11 altered to create various alternative effects levels by reducing the oxidized nitrogen loads by
12 rates of 5%, 10%, 20%, 30%, and 40% from their original 2002 levels. A zero percent reduction
13 corresponds to the 2002 current condition analysis. The remaining inputs to the SPARROW
14 model remained the same, and the model was rerun for each of these alternative effects level
15 scenarios. The total nitrogen load to the estuary calculated from the model was then converted to
16 TN_s using the annual average flow of the Potomac River. Plotting these concentrations against
17 the new total nitrogen atmospheric deposition loading (TN_{atm}) incorporating the oxidized
18 nitrogen reductions leads to the development of the first response curve and relationship (**Figure**
19 **5.2-14**) for the Potomac River and Potomac Estuary watershed.

20 For the second response curve, historical modeling data was used to determine total
21 nitrogen loads to the Potomac Estuary, which are then combined with annual average flow
22 values to calculate a final TN_s. These instream concentrations were then combined with the OEC
23 index scores, which were also determined from historical data, to create the data points needed to
24 create the 4-parameter logistic response curve in the *BackCalculation* program. **Figure 5.2-15**
25 presents an example of the logistic curve fit to the Potomac River and Potomac Estuary data
26 during an uncertainty analysis of a target ASSETS EI = 2.



1
2 **Figure 5.2-14.** Response curve relating instream total nitrogen concentration (TN_s) to
3 total nitrogen atmospheric deposition load (TN_{atm}) for the Potomac River watershed.



4
5 **Figure 5.2-15.** Example of fitted OEC curve for target ASSETS EI=2 for the Potomac
6 Estuary.

1 **Table 5.2-3** presents the
 2 summary statistics of 500
 3 iterations for each target
 4 ASSETS EI scenario for the
 5 Potomac Estuary. In these Monte
 6 Carlo type simulations, the TN_{atm}
 7 to the watershed is evaluated as a
 8 function of the TN_s for each step
 9 in improvement of the ASSETS
 10 EI.

11 The target ASSETS EI
 12 scenario where EI = 2

14 (improving from an ASSETS EI score of *Bad* to *Poor*) is
 16 the most interesting scenario and illustrates the power of
 18 the uncertainty analysis. The mean and median TN_{atm}
 20 values are negative, meaning again that not only must all

21 total nitrogen atmospheric deposition load (including all NO_x) be removed, but additional
 22 nitrogen from other sources must be removed as well. However, there is a slim chance that
 23 scenario ASSETS EI = 2 can be attained only from TN_{atm} deposition load reduction, as indicated
 24 by the positive 95th percentile TN_{atm} value of 9.02×10^6 (representing a 78% reduction).

25 Target scenario ASSETS EI = 3 (*Moderate*) is a unique case because all solutions were
 26 infeasible. With a TN_s value of 0 mg/L, the other (fixed) components of the ASSETS scoring
 27 methodology (i.e., DFO and Susceptibility Score) preclude satisfying any of the 95 combinations
 28 of DFO, OEC, and OHI that comprise the EI=3 combinations in the ASSETS lookup table.

29 Target scenario ASSETS EI = 4 and 5 had identical results. All 500 iterations returned a
 30 $TN_s^* = 0$, and a corresponding TN_{atm}^* negative load equal to $TN_{atm}^* = (0 - 2.72)/1.69 \times 10^{-8} = -$
 31 1.61×10^{-8} kg/yr. Clearly, target EIs equaling 4 and 5 are very much unattainable when reducing
 32 the total atmospheric nitrogen deposition load is the only policy option. To reach the target
 33 ASSETS EI scenario, total nitrogen atmospheric deposition (TN_{atm}) must be removed plus an
 34 additional amount (represented by the negative resultant load corresponding to $TN_s^* = 0$) that is
 35 approximately equal to one order of magnitude greater than the original atmospheric deposition

Table 5.2-3. Summary Statistics for Target
 Eutrophication Index Scenarios — Potomac Estuary
 (Current condition Eutrophication Index score of *Bad*)

Statistic	TN _{atm} (kg N/yr)	% TN _{atm} Reduction
ASSETS EI = 2 (<i>Poor</i>)		
Mean	-1.78×10^6	104
Median	-1.46×10^6	104
5th Percentile	-3.67×10^6	109
95th Percentile	9.02×10^6	78
ASSETS EI = 3 (<i>Moderate</i>)		
No feasible solutions found		
ASSETS EI=4 (<i>Good</i>) and ASSETS EI = 5 (<i>High</i>)		
All TN _{atm} = -1.61×10^8 , i.e., TN _s = 0 mg/L		

There is a slim chance that the Potomac River Estuary can move from an EI score of *Bad* to a score of *Poor* by reducing deposition of total nitrogen by 78%.

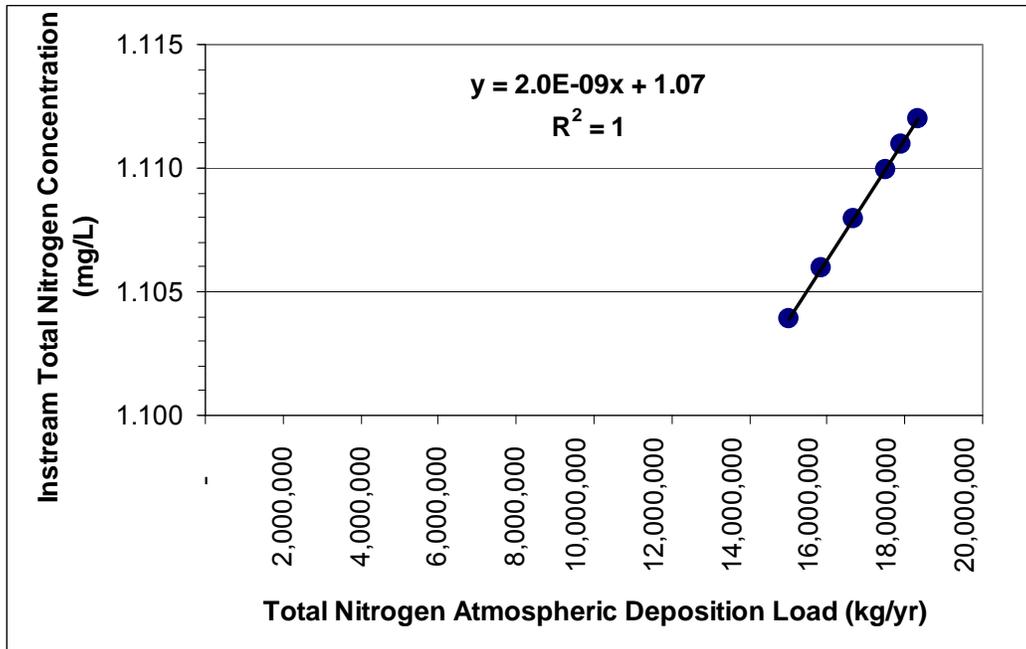
1 load. These amounts could be compared to the other nitrogen sources in the watershed (e.g.,
2 fertilizer and manure application or point sources) that were used as inputs to the SPARROW
3 model to determine the relative nature of the required removal with other sources in the
4 watershed. However, consideration must be given that this load is a reflection of the
5 characteristics of the source in the SPARROW model (e.g., spatial distribution, magnitude of
6 loads, sources/sinks), and a reduction required in atmospheric load is not equal to a reduction in
7 another source. Therefore, a reduction of an order of magnitude greater than the original
8 atmospheric deposition load may or may not be possible, depending on these different source
9 contributors. Relative proportions can be examined by comparing the source characteristics and
10 model parameters.

11 The SPARROW response curve can also be used to examine the role of atmospheric
12 nitrogen deposition in achieving specified reductions in total nitrogen estuarine load. For
13 example, the SPARROW modeling results predict that the 41×10^6 kg N/yr deposited
14 (atmospheric deposition input) over the Potomac River watershed in 2002 results in a loading of
15 7,380,000 kg N/yr, or 20% of the annual total nitrogen load, to the Potomac Estuary. If a 30%
16 reduction in annual total nitrogen load to the estuary (i.e., a reduction of 11×10^6 kg N/yr) were
17 desired, a reduction of 61×10^6 kg N/yr in nitrogen inputs to the watershed would be required
18 according to the SPARROW response curve based on atmospheric deposition. This represents a
19 100% reduction in the total nitrogen (including total reactive nitrogen) atmospheric deposition
20 inputs (41×10^6 kg N/yr) plus an additional 20×10^6 kg N/yr removal of nitrogen from other
21 sources in the Potomac River watershed (i.e., point and nonpoint sources). Note that this value of
22 20×10^6 kg N/yr is an approximate value when applied to the other sources because they differ
23 in characteristics (e.g., spatial distribution and magnitude) from atmospheric deposition that was
24 used to estimate the loading.

25 **5.2.7.2 Neuse River and Neuse River Estuary**

26 The same methods for creating alternative effects levels were applied to the data from the
27 Neuse River/Neuse River Estuary Case Study Area as to data from the Potomac River/Potomac
28 Estuary Case Study Area. The oxidized nitrogen atmospheric deposition loads were reduced by
29 rates of 5%, 10%, 20%, 30%, and 40% from their original 2002 levels. A zero percent reduction
30 corresponds to the 2002 current condition analysis. With the remaining inputs to the SPARROW

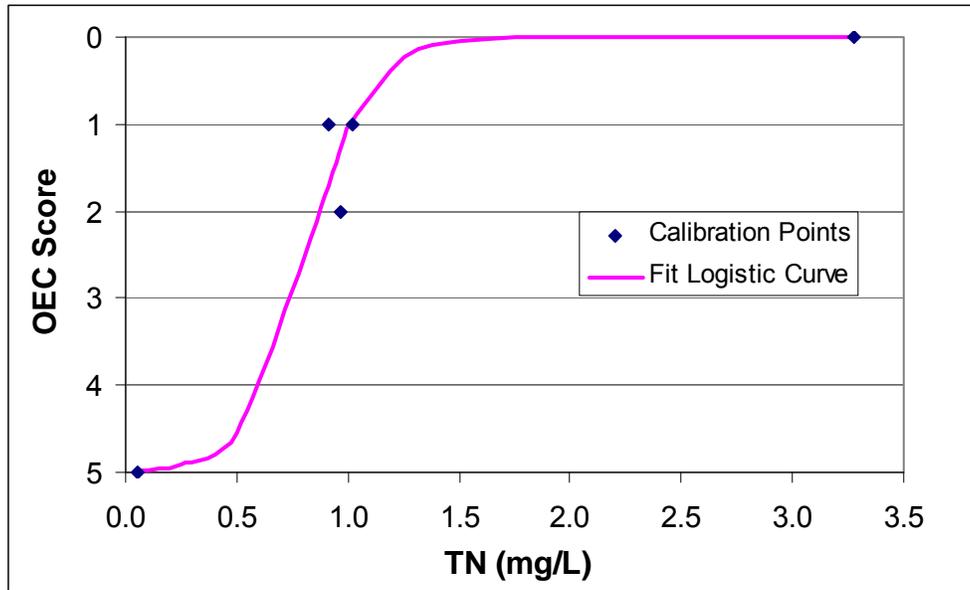
1 model kept the same, the SAS-developed model was rerun for each of these alternative effects
 2 level scenarios. The total nitrogen load to the estuary calculated from the model was then
 3 converted to an instream total nitrogen concentration (TN_s using the annual average flow of the
 4 Neuse River. Plotting these concentrations against the new total nitrogen atmospheric deposition
 5 load and incorporating the oxidized nitrogen reductions leads to the development of the first
 6 response curve and relationship (**Figure 5.2-16**).



7
 8 **Figure 5.2-16.** Response curve relating instream total nitrogen concentration to total
 9 nitrogen atmospheric deposition load for the Neuse River/Neuse River Estuary Case
 10 Study Area.

11 Historical monitoring data were used to determine instream total nitrogen concentrations
 12 at the downstream end of the Neuse River where the SPARROW model was used to determine
 13 current condition and alternative effects levels nitrogen loads. The monitoring data were derived
 14 from data downloaded from EPA's STORET Web site for monitoring location J8290000 from
 15 the North Carolina Division of Water Quality. These TN_s were then combined with the OEC
 16 index scores, which were also determined from historical data, to create the data points needed to
 17 create the logistic response curve in the *BackCalculation* program. Data from as many years as
 18 possible were gathered for both the TN_s and OEC index scores. However, due to the limited
 19 amount of complete data from the various sources identified under the current condition analysis,
 20 only three corresponding years of data were found. The theoretical ecological endpoints of the

1 curve were also used to create the 4-parameter logistic curve representing the second response
 2 curve for the analysis (**Figure 5.2-17**).



3
 4 **Figure 5.2-17.** Example of fitted response curve for target ASSETS EI=2 for the Neuse
 5 River Estuary.

6 Each of the four ASSETS EI scores representing state improvements (*Poor-2, Moderate-*
 7 *3, Good-4, High-5*) was treated as a “target” ASSETS EI score, and 500 Monte Carlo simulations
 8 were run under each target ASSETS EI scenario to relate instream total nitrogen concentrations
 9 (TN_s) to total nitrogen

10 atmospheric deposition (TN_{atm}).
 11 **Table 5.2-4.** Summary Statistics for Target
 12 Eutrophication Index Scenarios — Neuse River Estuary
 13 (Current condition Eutrophication Index score of *Bad*)

14 The summary statistics of
 15 the 500 iterations for each target
 16 ASSETS EI scenario are
 17 presented in **Table 5.2-4**.

18 For target scenario
 19 ASSETS EI = 2 (improving from
 20 an ASSETS EI score of *Bad* to
 21 *Poor*), all reductions exceed
 100%, meaning that not only
 must all TN_{atm} deposition load be
 removed to meet ASSETS EI = 2, but considerably more nitrogen from other sources as well.

Statistic	TN_{atm} (kg N/yr)	% TN_{atm} Reduction
ASSETS EI = 2 (<i>Poor</i>)		
Mean	-1.43×10^8	880
Median	-1.43×10^8	880
5th Percentile	-1.47×10^8	901
95th Percentile	-1.01×10^8	653
ASSETS EI = 3 (<i>Moderate</i>)		
No feasible solutions found		
ASSETS EI=4 (<i>Good</i>) and ASSETS EI = 5 (<i>High</i>)		
All $TN_{atm} = -5.35 \times 10^8$, i.e. $TN_s = 0$ mg/L		

1 Given these results, the Neuse River Estuary is currently somewhere between these two ASSETS
 2 EI values (*Bad* and *Poor*) as was the Potomac Estuary. There is some evidence that it is slightly
 3 more eutrophic than the Potomac Estuary, because there was at least a slim chance for the
 4 Potomac Estuary (at the 95th percentile) that TN_{atm} reductions (of <100%) would achieve
 5 ASSETS EI = 2.

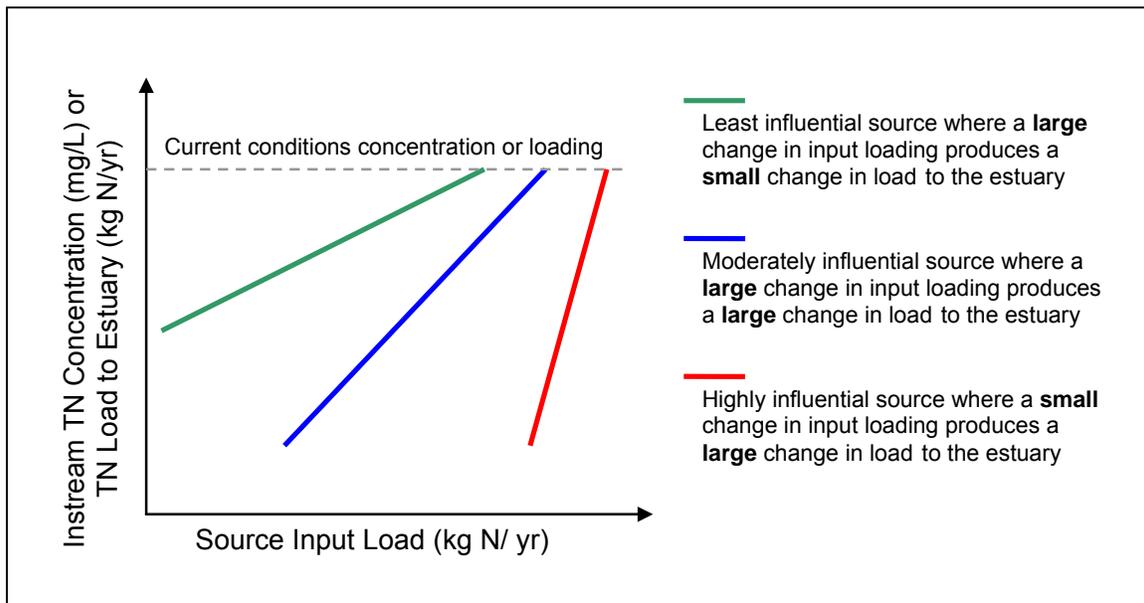
6 Target scenario ASSETS EI = 3 (*Moderate*) is again a unique case because all solutions
 7 were infeasible as described above for the Potomac River watershed and the Potomac Estuary.
 8 (See Appendix 6 for further explanation.)

9 Target scenarios ASSETS EI = 4 (*Good*) and 5 (*High*) had identical results. All 500
 10 iterations returned a $TN_s = 0$, and a corresponding TN_{atm} negative load equal to $TN_{atm} = (0 -$
 11 $1.07)/2.0 \times 10^{-9} = -5.35 \times 10^8$ kg/yr. Clearly, target scenarios of ASSETS EI equal 4 (*Good*) and 5
 12 (*High*) are unattainable when reducing the TN_{atm} (including all NO_x) is the only option. Again,
 13 the reduction required includes all of the total reactive nitrogen atmospheric deposition source
 15 plus a load an order of magnitude greater than the original atmospheric deposition load (10^8
 17 kg/yr), which could be compared to the other
 19 nitrogen sources used as inputs to the SPARROW
 21 model giving consideration to the characteristics
 23 of each of these sources. Again, this additional
 24 magnitude of reduction may or may not be feasible based on the relative contributions from the
 25 other sources.

To change the Neuse River Estuary's EI score from *Bad* to *Poor*, not only must 100% of the total atmospheric nitrogen deposition be eliminated, but considerably more nitrogen from other sources as well must be reduced.

26 The SPARROW response curve can be used to examine the role of atmospheric nitrogen
 27 deposition in achieving desired reductions load to the Neuse River Estuary. In the Neuse River
 28 watershed, modeling results indicate that 18×10^6 kg N/yr was deposited in 2002. SPARROW
 29 modeling predicts that this deposition input results in a loading of 1.2×10^6 kg N/yr (26% of the
 30 annual total nitrogen load) to the Neuse River Estuary. Unlike the Potomac River and Potomac
 31 Estuary, little change is seen in the total nitrogen loading to the Neuse River Estuary, with large
 32 decreases in the nitrogen deposition. If all atmospheric nitrogen deposition inputs were
 33 eliminated (100% reduction), the total annual nitrogen load to the Neuse River Estuary would
 34 only decrease by 4%. This small effect is because the total nitrogen loadings to the Neuse River
 35 Estuary are so dependent on the other sources within the SPARROW model. That is, the
 36 SPARROW response curve cannot be used to predict the relative magnitudes of loads needed to

1 produce reductions greater than this 4%. This lack of predictive power of the response curve
 2 based on atmospheric deposition is due to the differences in characteristics between the sources
 3 within the watershed, where fertilizer, in particular, has a strong signature (i.e., indicating the
 4 large influence of agriculture within the watershed). This result shows that the SPARROW
 5 response curves of total nitrogen load to other sources would be quite different. **Figure 5.2-18**
 6 illustrates the theoretical response curves that may result when the SPARROW modeled loads
 7 are plotted against the other total nitrogen source inputs. The green curve, or least influential
 8 source, displays the behavior of the atmospheric deposition for the Neuse River Estuary. The red
 9 curve, or highly influential source, likely corresponds to how agricultural sources within the
 10 watershed behave. These response curves will depend on the source magnitudes, spatial
 11 distributions, and other characteristics.



12
 13 **Figure 5.2-18.** Theoretical SPARROW response curves demonstrating relative influence
 14 of sources on nitrogen loads to an estuary.

15 **5.2.8 Uncertainty and Variability**

16 The use of multiple datasets, predictive modeling, and a multi-indicator assessment tool,
 17 such as ASSETS, requires consideration of the impact of data variability and the uncertainties in
 18 case study results.

19 “Uncertainty is a measure of the knowledge of the magnitude of a parameter. Uncertainty
 20 can be reduced by research, i.e., the parameter value can be refined. Uncertainty is quantified as
 21 a distribution. For example, the volume of a lake may be estimated from its surface area and an

1 average depth. This estimate can be refined by measurement. Variance is a measure of the
2 heterogeneity of a landscape parameter or the inherent variability in a chemical property.
3 Variance cannot be reduced by further research. It is quantified as a distribution. For example,
4 the organic carbon content of the soil in a region may vary, even over short distances. The soil is
5 not homogenous and thus the organic carbon content can be described with a distribution of
6 values.” (Webster and MacKay, 2003)

7 Uncertainty with this method of assessment for aquatic nutrient enrichment may include
8 the following:

- 9 ▪ **Data inputs to SPARROW.** For this study, the data used were developed under separate
10 studies and published by the USGS. Because the data were independently verified before
11 publication by the USGS, only quality checks were performed on the data, rather than full
12 validation exercises.
- 13 ▪ **Modeling uncertainty in SPARROW estimates.** The Version 3 Chesapeake Bay
14 SPARROW application met evaluation criteria based on degrees of freedom, model error,
15 and R squared values. The calibration of the Neuse watershed SPARROW model using
16 SAS examined the standard deviation, t-statistics, p-values, and Variance Inflation Factors
17 (VIF) for each estimated parameter. The model derived for the Neuse River watershed did
18 produce some model parameters (e.g., manure production, urban area, decay terms) that
19 did not reach desired statistical significance levels.
- 20 ▪ **Sensitivity of SPARROW formulation due to atmospheric inputs in the Aquatic**
21 **Nutrient Enrichment Case Study.** While it is certain that the parameter estimated to
22 apply to the atmospheric deposition source will change, what is uncertain at this point is
23 the extent to which the other model parameters and the overall nitrogen load estimates will
24 be affected by using the CMAQ/NADP estimates in the model calibrated against the wet
25 nitrate deposition values.
- 26 ▪ **Calibration data for SPARROW estimates.** Monitoring data were used to calibrate the
27 SPARROW model. By relying on data from federally recognized data systems, the aim is
28 to use data that has undergone quality assurance/quality control procedures. Additionally,
29 collaboration has been completed with the researchers who have conducted the previous
30 SPARROW applications in each case study area to provide a rigorous check on the data
31 used.

- 1 ▪ **Data inputs to the ASSETS EI.** Because of the numerous data requirements and sources
2 required to conduct a full ASSETS EI analysis, there is a large range of uncertainty that
3 can enter into the calculations. Best attempts were made to apply standardized evaluation
4 methods in order to minimize any uncertainties due to subjectivity or processing
5 differences.
- 6 ▪ **Heuristic estimates of future outlook.** The estimation of the future outlook score in the
7 ASSETS EI currently relies on heuristic estimates from systems experts.
- 8 ▪ **Steady-state estimates/mean annual estimates.** Both SPARROW and the ASSETS EI
9 currently provide only longer-term estimates of the system conditions.
- 10 ▪ **Use of a Screening Method.** The methods used in this study are only of the screening
11 level. The screening method provides a response curve that can be used in the evaluation
12 of ecosystem services. Additionally, many of the complex concepts linking the indicators
13 of eutrophication to the effects of eutrophication are not highly developed or understood at
14 this time (Howarth and Marino, 2006).

15 Uncertainties in the Back Calculation Methods include the following:

- 16 ▪ **Missing ASSETS EI rankings per combinations of index scores.** The combinations of
17 OHI, OEC, and DFO scores provided by Bricker et al. (2003) leave out 30 of the possible
18 125 combinations that represent overall ASSETS scores.
- 19 ▪ **Better rationale for TN minimum and maximum uncertainty range.** The assigned
20 uncertainty ranges were based on best professional judgment, but more research is needed.
21 The results presented herein for the Potomac and Neuse River estuaries should be
22 interpreted as illustrative of the methodology, not strictly valid.
- 23 ▪ **Methodology to incorporate uncertainty in the SPARROW model.** Estimates of TN_s at
24 the head of the estuary, predicted by SPARROW and driven by the TN_{atm} (i.e., total
25 nitrogen deposition evaluated on reductions in NO_x) over the watershed and other nitrogen
26 sources, are uncertain. That uncertainty was not considered in these two case studies;
27 therefore, the probability distributions of TN_{atm}^* presented are artificially “tight” (i.e., the
28 true distributions would exhibit more variability).
- 29 ▪ **More convergence testing to determine appropriate numbers of samples.** Some
30 modest convergence testing was completed to determine how many samples of the

OEC(TN) function need to be used in order for the statistics of interest for the resulting NO_xi^* distributions to be reasonably stable. More convergence testing is needed.

- **Crossing of a categorical ranking system with a continuous nitrogen concentration scale.** Several assumptions and considerations had to be made in order to create and evaluate the logistic response curve because the OEC index score is a categorical ranking of 1 through 5, whereas TN_s is a continuous variable. The functions evaluated in *BackCalculation* treat the OEC index score as a continuous function. Until higher-level models are developed to relate the nitrogen concentrations in the system to eutrophication effects, these assumptions are necessary. Future applications with additional data should be used to test and validate these assumptions and results.

5.3 TERRESTRIAL NUTRIENT ENRICHMENT

Terrestrial nutrient enrichment is described in the ISA (U.S. EPA, 2008, Section 3.3) for many different ecosystems. In particular, additional nitrogen may affect the plants in these ecosystems. Changes to the individual plants as well as changes to populations and communities of plants have been documented. Over the last half century, landscapes in the United States have been exposed to atmospherically deposited nitrogen from anthropogenic activities. Some of the highest nitrogen deposition has occurred in Southern California, where researchers have documented measurable ecological changes related to atmospheric deposition. Evidence from the two ecosystems discussed in this case study—CSS and MCF communities in the Sierra Nevada Range and San Bernardino Mountains of California—supports the finding that nitrogen alters these habitats. Changes in nitrogen loading may also affect the ecological services provided by the CSS and MCF ecosystems, including regulation (e.g., water, habitat), cultural and aesthetic value (e.g., recreation, natural landscape, sense of place), and provisioning (e.g., timber) (MEA, 2005). The Terrestrial Nutrient Enrichment Case Study also evaluated research conducted on these complex ecosystems to understand the relationships among the effects of nitrogen loads, fire frequency and intensity, and invasive plants.

Section 3.3 of the ISA (U.S. EPA, 2008) describes the ecosystems and species of concern, identifies trends in the ecosystems and the effects of these trends, and discusses research efforts that investigated the variables and driving forces that may affect the communities. The CMAQ 2002 modeling results and the NADP monitoring data for 2002 were

1 used to gain an understanding of how atmospheric deposition of nitrogen is spatially distributed.
2 GIS data on the spatial extent of the habitat and associated habitat changes, the location of fire
3 threat, and the location of sensitive species were used to compare these patterns to the
4 CMAQ/NADP data. In sum, spatial information and observed, experimental effects were used to
5 help identify the trends in these ecosystems and to describe the past and current spatial extent of
6 the ecosystems.

7 Current analysis of the effects of terrestrial nutrient enrichment from atmospheric
8 nitrogen deposition in both CSS and MCF ecosystems seeks to improve scientific understanding
9 of the interactions among nitrogen deposition, fire events, and community dynamics. The
10 available scientific information is sufficient to identify ecological benchmarks that are affected
11 by nitrogen deposition. Ecological benchmarks have been identified for CSS and MCF.

12 **5.3.1 Ecological Indicators, Ecological Responses, and Ecosystem Services**

13 **5.3.1.1 Indicators**

14 Ecosystems may respond to the addition of nitrogen in a number of ways. There may be
15 gains in productivity and growth initially. Increasing levels of nitrogen; however, may lead to
16 changes in community structure and function, with changes in species composition or changes in
17 the abundance and distribution of organisms. If changes include loss of threatened, endangered
18 or rare species, or rare communities or a diminished productivity or increased fire threat, then
19 such changes would be cause for concern. Indicators of possible changes can be identified that
20 would assist in determining an acceptable ambient air concentration of nitrogen oxides.

21 Terrestrial nutrient enrichment research has measured ecosystems' exposure to deposition of
22 various atmospheric nitrogen species, including nitrogen oxides, reduced nitrogen, and total
23 nitrogen. The ISA (U.S. EPA, 2008, Section 3.3) documents current understanding of the effects
24 of nitrogen nutrient enrichment on terrestrial ecosystems. The ISA concludes that there is
25 sufficient information to infer a causal relationship between atmospheric nitrogen deposition and
26 biogeochemical cycling and fluxes of nitrogen in terrestrial systems. The ISA further concludes
27 that there is a causal relationship between atmospheric nitrogen deposition and changes in
28 species richness, species composition, and biodiversity in terrestrial systems. These conclusions
29 are based on an extensive literature review that is summarized in Table 4-4 of the ISA. The
30 science review includes both observational and experimental (nitrogen addition) research. Alpine

1 ecosystems, grasslands (including arid and semiarid ecosystems), forests, and deserts were
2 included. This extensive documentation was used to assist in selecting the case study sites to
3 identify and compare ecological benchmarks from different ecosystems.

4 CSS is subject to several pressures, such as land conversion, grazing, fire, and pollution,
5 all of which have been observed to induce declines in other ecosystems (Allen et al., 1998).
6 Research has shown that both fire and increased nitrogen can enhance the growth of nonnative
7 grasses in established CSS communities (Keeley et al., 2005; Wood et al., 2006). It is
8 hypothesized that many CSS stands are no longer limited by nitrogen and have instead become
9 nitrogen-saturated because of atmospheric nitrogen deposition (Allen et al., 1998; Westman,
10 1981). Nitrogen availability may favor the germination and growth of nonnative grasses, which
11 can create a dense network of shallow roots that slow the diffusion of water through soil,
12 decrease the percolation depth of precipitation, and decrease the water storage capability of the
13 soil and underlying bedrock (Wood et al., 2006). CSS has been declining in land area and in
14 shrub density for the past 60 years, and in many places it is being replaced by nonnative annual
15 grasses (Allen et al., 1998; Padgett and Allen, 1999). Replacement by nonnative grasses results
16 in less habitat for threatened and endangered species and also appears to increase fire
17 vulnerability. Atmospheric nitrogen deposition has been suggested as a possible cause or factor
18 in this ecosystem alteration (U.S. EPA 2008, Section 3.3). Changes in community metrics may,
19 therefore, be useful indicators of atmospheric nitrogen deposition for CSS.

20 The ISA discusses the extensive land areas in the western United States that receive low
21 levels of atmospheric nitrogen deposition and that are interspaced with areas of relatively higher
22 atmospheric deposition downwind of large metropolitan centers and agricultural areas. Fenn et
23 al. (2008) determined empirical critical loads (i.e., measured levels of nitrogen at a specific
24 location where biological impacts occur) for atmospheric nitrogen deposition in MCF based on
25 changes in leached nitrate in receiving waters, reduced fine-root biomass in Ponderosa pine
26 (*Pinus ponderosa*), and epiphytic lichen communities. Lichens are good early indicators of
27 atmospheric nitrogen deposition effects on other MCF species because lichens rely entirely on
28 atmospheric nitrogen and cannot regulate uptake. (Figure 5.3-1)

1 From the lichen data, Fenn et
 2 al. (2008) predicted that a critical
 3 load of 3.1 kg N/ha/yr would be
 4 protective for all components of the
 5 forest ecosystem. The study further
 6 found that an atmospheric nitrogen
 7 deposition of 17 kg N/ha/yr was
 8 associated with NO_3^- leaching and an
 9 approximately 25% reduction in fine
 10 root biomass.

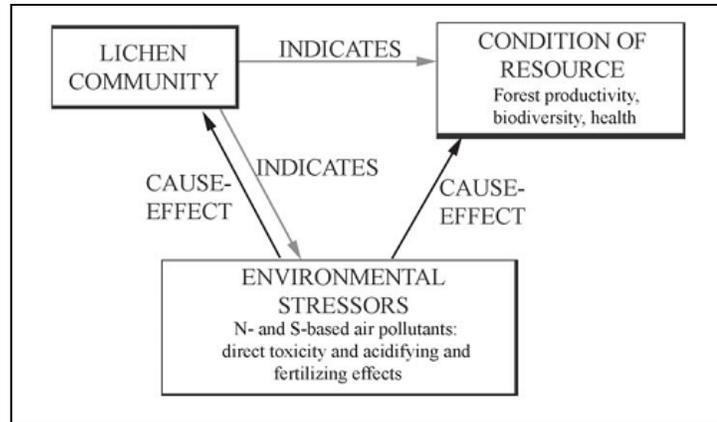


Figure 5.3-1. Importance of lichens as an indicator of ecosystem health (Jovan, 2008).

11 5.3.1.2 Ecological Responses: Benchmark Values Selected for This Case Study

12 The data limitations on atmospheric nitrogen deposition described above, along with
 13 current data to describe the full extent and distribution of nitrogen sensitive U.S. ecosystems,
 14 presented a barrier to designing a case study that uses quantitative monitoring and modeling
 15 tools. Instead, this case study used published research results to identify meaningful ecological
 16 benchmarks associated with different levels of atmospheric nitrogen deposition.

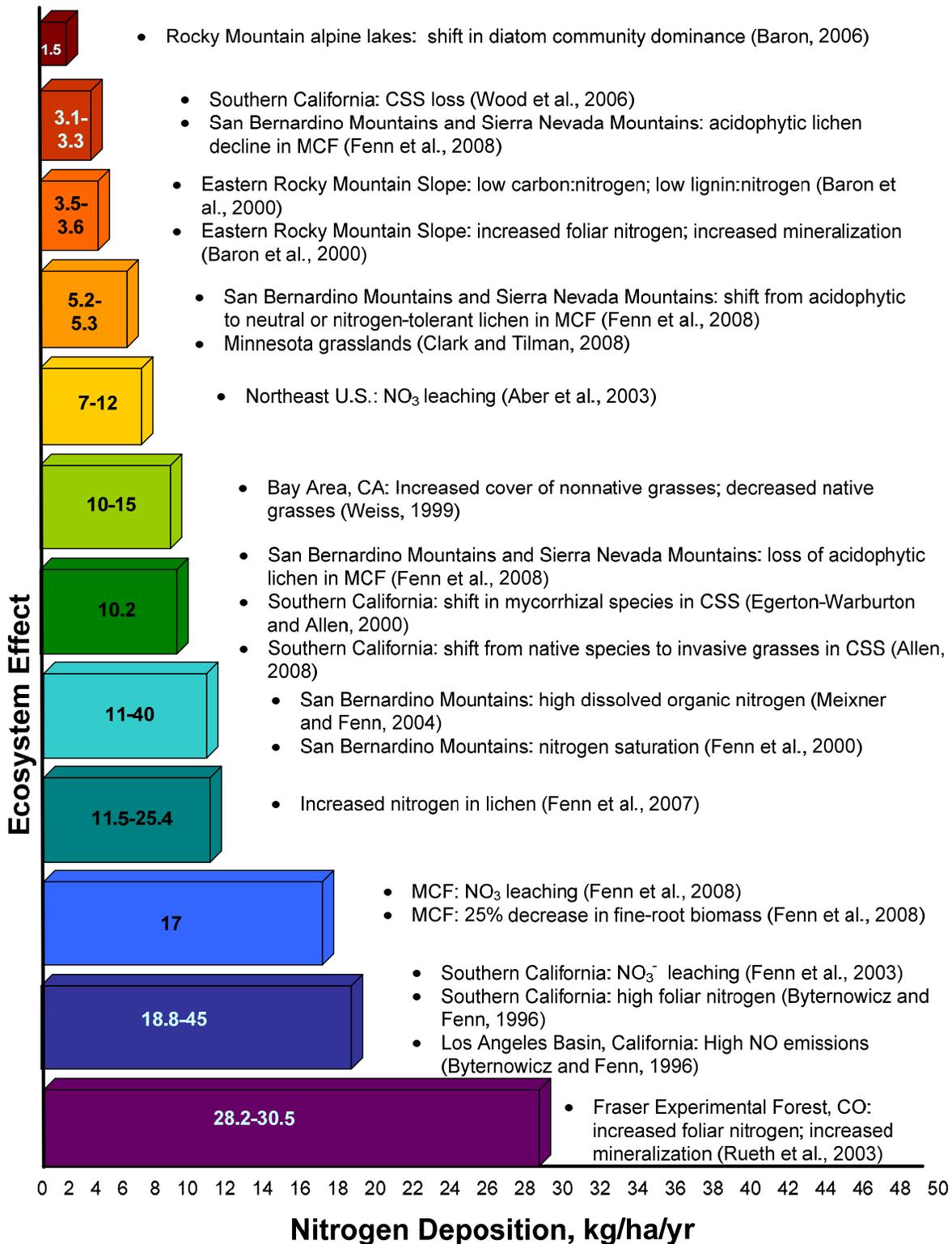
17 The ecological benchmarks that were identified for the CSS and the MCF are included in
 18 the suite of benchmarks identified in the ISA (U.S. EPA, 2008, Section 3.3). There are sufficient
 19 data to confidently relate the ecological effect to a loading of atmospheric nitrogen. For the CSS
 20 community, the following ecological benchmarks were identified:

- 21 ▪ 3.3 kg N/ha/yr – the amount of nitrogen uptake by a vigorous stand of CSS; above this
 22 level, nitrogen may no longer be limiting
- 23 ▪ 10 kg N/ha/yr – mycorrhizal community changes

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

- 25 ▪ 3.1 kg N/ha/yr – shift from sensitive to tolerant lichen species
- 26 ▪ 5.2 kg N/ha/yr – dominance of the tolerant lichen species
- 27 ▪ 10.2 kg N/ha/yr – loss of sensitive lichen species
- 28 ▪ 17 kg N/ha/yr – leaching of nitrate into streams.

29 These benchmarks, as well as those from other systems, are presented in **Figure 5.3-2**.



1
2 **Figure 5.3-2.** Benchmarks of atmospheric nitrogen deposition for several ecosystem indicators.

5.3.1.3 Ecosystem Services

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

Both CSS and MCF are located in areas of California valuable for housing, recreation, and development. CSS runs along the coast through densely populated areas of California. MCF covers less densely populated areas that are valuable for recreation. (Appendix 8, Figure 5.1-1) The proximity of CSS and MCF to population centers and recreational areas and the potential value of these landscape types in providing regulating ecosystem services suggest that the value of preserving CSS and MCF to California could be quite high. The value that California residents and the U.S. population as a whole place on CSS and MCF habitats is reflected in the various federal, state, and local government measures that have been put in place to protect these habitats. Threatened and endangered species are protected by the Endangered Species Act. The State of California passed the Natural Communities Conservation Planning Program in 1991, and CSS was the first habitat identified for protection under the program (see <http://www.dfg.ca.gov/habcon/nccp/>). Private organizations, such as The Nature Conservancy, the Audubon Society, and local land trusts also protect and restore CSS and MCF habitats. According to the 2005 National Land Trust Census Report (Land Trust Alliance, 2006), California has the most land trusts of any state, with a total of 1,732,471 acres either owned, under conservation easement, or conserved by other means.

Cultural

The primary cultural ecosystem services associated with CSS and MCF are recreation, aesthetic, and nonuse values. The possible ecosystem service benefits from reducing nitrogen enrichment in CSS and MCF and a general overview of the types and relative magnitude of the benefits are discussed below.

CSS, once the dominant landscape type in the area, is a unique ecosystem that provides cultural value to California and the nation as a whole. Culturally, the remaining patches of CSS

1 contain a number of threatened and endangered species, and patches of CSS are present in a
2 number of parks and recreational areas. More generally, the patches of CSS represent the iconic
3 landscape of Southern California and serve as a reminder of what the area looked like pre-
4 development. Changes that might impact cultural ecosystem services in CSS resulting from
5 nutrient enrichment potentially include the following:

- 6 ▪ Decline in CSS habitat, shrub abundance, and species of concern
- 7 ▪ Increased abundance of nonnative grasses and other species
- 8 ▪ Increase in wildfires.

9 For MCF, the changes from nutrient enrichment that might impact cultural ecosystem
10 services include the following:

- 11 ▪ Change in habitat suitability and increased tree mortality
- 12 ▪ Decline in MCF aesthetics.

13 **Recreation**

14 CSS and MCF are found in numerous recreational areas in California. Three national
15 parks and monuments in California contain CSS, including Cabrillo National Monument,
16 Channel Islands National Park, and Santa Monica National Recreation Area. All three parks
17 showcase CSS habitat with educational programs and information provided to visitors, guided
18 hikes, and research projects focused on understanding and preserving CSS. A total of 1,456,879
19 visitors traveled through these three parks in 2008. MCF is highlighted in Sequoia and Kings
20 Canyon National Park, Yosemite National Park, and Lassen Volcanic National Park, where a
21 total of 5,313,754 people visited in 2008. In addition, numerous state and county parks
22 encompass CSS and MCF habitat. For example, California’s Torrey Pines State Natural Reserve
23 protects CSS habitat (see <http://www.torreypine.org/>). Visitors to these parks engage in activities
24 such as camping, hiking, attending educational programs, horseback riding, wildlife viewing,
25 water-based recreation, and fishing.

26 The 2006 FHWAR for California (U.S. DOI, 2007) reports on the number of individuals
27 involved in fishing, hunting, and wildlife viewing in California. Millions of people are involved
28 in these three activities each year. The quality of these trips depends in part on the health of the
29 ecosystems and their ability to support the diversity of plants and animals found in important
30 habitats. Based on estimates from Kaval and Loomis (2003), in the Pacific Coast region of the

1 United States, a day of fishing has an *average* value of \$48.86 (in 2007 dollars) based on 15
2 studies. For hunting and wildlife viewing in this region, average day values were estimated to be
3 \$50.10 and \$79.81 from 18 and 23 studies, respectively. Multiplying these average values by the
4 total participation days reported in Appendix 8, **Table 5.1-1**, the total benefits in 2006 from
5 fishing, hunting, and wildlife viewing away from home in California were approximately \$947
6 million, \$169 million, and \$3.59 billion, respectively.

7 In addition, data from California State Parks (2003) indicate that in 2002, 68.7% of adult
8 residents participated in trail hiking, for an average of 24.1 days per year. Applying these same
9 rates to Census estimates of the California adult population in 2007 suggests that there were
10 roughly 453 million days of hiking by residents in California in 2007. According to Kaval and
11 Loomis (2003), the average value of a hiking day in the Pacific Coast region is \$25.59, based on
12 a sample of 49 studies. Multiplying this average day value by the total participation estimate
13 indicates that the aggregate annual benefit for California residents from trail hiking in 2007 was
14 \$11.59 billion.

15 **Aesthetic**

16 Beyond the recreational value, the CSS landscape and MCF provide aesthetic services to
17 local residents and homeowners who live near CSS or MCF. Aesthetic services not related to
18 recreation include the view of the landscape from houses, as individuals commute, and as
19 individuals go about their daily routine in a nearby community. Studies find that scenic
20 landscapes are capitalized into the price of housing. While there are no known studies that look
21 at the value of housing as a function of the view in landscapes that include CSS or MCF, other
22 studies document the existence of housing price premia associated with proximity to forest and
23 open space (Acharya and Bennett, 2001; Geoghegan et al., 1997; Irwin, 2002; Mansfield, et al.,
24 2005; Smith et al., 2002; Tyrvaainen and Miettinen, 2000). The CSS landscape itself is closely
25 associated with Southern California, which should increase the aesthetic value of the landscape
26 in general. CSS areas border a number of areas along the coast near large cities with very high
27 home values, as well as areas between the cities where home values are lower.

28 **Nonuse Value**

29 Nonuse value, also called existence value or preservation value, encompasses a variety of
30 motivations that lead individuals to place value on environmental goods or services that they do
31 not use. The values individuals place on protecting rare species, rare habitats, or landscape types

1 that they do not see or visit and that do not contribute to the pleasure they get from other
2 activities are examples of nonuse values.

3 While measuring the public’s willingness to pay to protect endangered species poses
4 theoretical and technical challenges, it is clear that the public places a value on preserving
5 endangered species and their habitats. Data on charitable donations, survey results, and the time
6 and effort different individuals or organizations devote to protecting species and habitats suggest
7 that endangered species have intrinsic value to people beyond the value derived from using the
8 resource (e.g., recreational viewing or aesthetic value). CSS and MCF are home to a number of
9 important and rare species and habitat types. CSS displays richness in biodiversity, with more
10 than 550 herbaceous annual and perennial species. Of these herbs, nearly half are endangered,
11 sensitive, or of special status (Burger et al., 2003). Additionally, avian, arthropod, herpetofauna,
12 and mammalian species live in CSS habitat or use the habitat for breeding or foraging.

13 Communities of CSS are home to three important federally endangered species. MCF is
14 home to one federally endangered species and a number of state-level sensitive species. The
15 Audubon Society lists 28 important bird areas in CSS habitat and at least 5 in MCF in California
16 (<http://ca.audubon.org/iba/index.shtml>).³

17 Only one known study has specifically estimated values for protecting CSS habitat in
18 California. Stanley (2005) uses a contingent valuation (CV) survey to measure willingness to pay
19 (WTP) to support recovery plans for endangered species in Southern California. The survey of
20 Orange County, CA, residents asked respondents to value the recovery of a single species (i.e.,
21 the Riverdale fairy shrimp) and a larger bundle of 32 species found in the county. The
22 acquisition of critical habitat and implementation of the recovery plan were the specific goods
23 being valued in the WTP question, and the programs would be financed by an annual tax
24 payment. The average WTP for Riverdale fairy shrimp recovery was roughly \$29 (in 2007
25 dollars), and for all 32 species, it was \$61 per household, depending on the model used.
26 Aggregating benefits (i.e., multiplying average household WTP by the number of households in
27 the county) results in total estimated WTP of more than \$27 million annually for protecting
28 Riverdale fairy shrimp and \$57 million annually for protecting all 32 species.

29 In a more general study valuing endangered species protection, Loomis and White (1996)
30 synthesize key results from 20 threatened and endangered species valuation studies using meta-

³ Important Bird Areas are sites that provide essential habitat for one or more species of bird.

1 analysis methods. They find that annual WTP estimates range from a low of \$11 for the Striped
2 Shiner fish to a high of \$178 for the Northern Spotted Owl (in 2007 dollars). None of the studies
3 summarized by Loomis and White are found in CSS or MCF, but the study provides another
4 indication of the value that the public places on preserving endangered species in general.

5 **Regulating**

6 Excessive nitrogen deposition upsets the balance between CSS and nonnative plants,
7 changing the ability of an area to support the biodiversity found in CSS. The composition of
8 species in CSS changes fire frequency and intensity, as nonnative grasses fuel more frequent and
9 more intense wildfires. More frequent and intense fires also reduce the ability of CSS to
10 regenerate after a fire and increase the proportion of nonnative grasses (U.S. EPA, 2008). A
11 healthy MCF ecosystem supports native species, promotes water quality, and helps regulate fire
12 intensity. Excess nitrogen deposition leads to changes in the forest structure, such as increased
13 density and loss of root biomass, which, in turn, can result in more intense fires and water quality
14 problems related to nitrate leaching (U.S. EPA, 2008).

15 The importance of CSS and MCF as homes for sensitive species and their aesthetic
16 services have been discussed in Appendix 8, Section 5.1.1. Here the contribution of CSS and
17 MCF to fire regulation and water quality is discussed.

18 **Fire Regulation**

19 The terrestrial enrichment case study identified fire regulation as a service that could be
20 affected by nutrient enrichment of the CSS and MCF ecosystems by encouraging growth of more
21 flammable grasses. Wildfires represent a serious threat in California and cause billions of dollars
22 in damage. Over the 5-year period from 2004 to 2008, Southern California experienced, on
23 average, more than 4,000 fires per year burning, on average, more than 400,000 acres per year
24 (NASF, 2009). Improved fire regulation leads to short-term and long-term benefits. The short-
25 term benefits include the value of avoided residential property damages, avoided damages to
26 timber, rangeland, and wildlife resources, avoided losses from fire-related air quality
27 impairments, avoided deaths and injury due to fire, improved outdoor recreation opportunities,
28 and savings in costs associated with fighting the fires and protecting lives and property. For
29 example, the California Department of Forestry and Fire Protection (CAL FIRE) estimated that
30 average annual losses to homes due to wildfire from 1984 to 1994 were \$163 million per year
31 (CAL FIRE, 1996) and were more than \$250 million in 2007 (CAL FIRE, 2008). In fiscal year

1 2008, CAL FIRE’s costs for fire suppression activities were nearly \$300 million (CAL FIRE,
2 2008). Therefore, even a 1% reduction in these damages and costs would imply benefits of more
3 than \$5 million per year.

4 CSS overlaps with areas of very high to extremely high fire threat. MCF is found in some
5 areas closer to the coast with extremely high fire threat and in areas in the mountains also under
6 very high fire threat.

7 In the long term, decreased frequency of fires could result in an increase in property
8 values in fire-prone areas. Mueller et al. (2007) conducted a hedonic pricing study to determine
9 whether increasing numbers of wildfires affect house prices in Southern California. They
10 estimated that house prices would decrease 9.71% (\$30,693 in 2007 dollars) after one fire and
11 22.7% (\$71,722; \$102,417 cumulative) after a second wildfire within 1.75 miles of a house in
12 their study area. After the second fire, the housing prices took between 5 and 7 years to recover.
13 The results come from a sample of 2,520 single-family homes located within 1.75 miles of one
14 of five fires during the 1990s.

15 Long-term decreases in wildfire risks are also expected to provide outdoor recreation
16 benefits. The empirical literature contains several articles measuring the relationship between
17 wildfires and recreational values; however, very few address fires in California, particularly in
18 CSS areas. One exception is Loomis et al. (2002), which estimates the changes in deer harvest
19 and deer hunting benefits resulting from controlled burns or a prescribed fire in the San
20 Bernardino National Forest in Southern California. Using a CV survey of deer hunters in
21 California, they estimated that the net economic value of an additional deer harvested is on
22 average \$122 (in 2007 dollars). Based on predicted changes in deer harvest in response to a
23 prescribed fire, they estimated annual economic benefits for an additional 1,000 acres of
24 prescribed burning ranges from \$3,328 to \$3,893.

25 **Water Quality**

26 In the MCF Case Study, maintaining water quality emerged as a regulating service that
27 can be upset by excessive nitrogen. When the soil becomes saturated, nitrates may leach into the
28 surface water and cause acidification. Several large rivers and Lake Tahoe cut through MCF
29 areas (see Appendix 8, **Figure 5.1-10**). Additional nitrogen from MCF areas could further
30 degrade waters that are already stressed by numerous other sources of nutrients and pollution.

31 **Value of Coastal Sage Scrub and Mixed Conifer Forest Ecosystem Services**

1 The CSS and MCF were selected as case studies for terrestrial nutrient enrichment
 2 because of the potential that these areas could be adversely affected by excessive nitrogen
 3 deposition. To date, the detailed studies needed to identify the magnitude of the adverse impacts
 4 due to nitrogen deposition have not been completed. Based on available data, this *Risk and*
 5 *Exposure Assessment* report provides a qualitative discussion of the services offered by CSS and
 6 MCF and a sense of the scale of benefits associated with these services. California is famous for
 7 its recreational opportunities and beautiful landscapes. CSS and MCF are an integral part of the
 8 California landscape, and together the ranges of these habitats include the densely populated and
 9 valuable coastline and the mountain areas. Through recreation and scenic value, these habitats
 10 affect the lives of millions of California residents and tourists. Numerous threatened and
 11 endangered species at both the state and federal levels reside in CSS and MCF. Both habitats
 12 may play an important role in wildfire frequency and intensity, an extremely important problem
 13 for California. The potentially high value of the ecosystem services provided by CSS and MCF
 14 justify careful attention to the long-term viability of these habitats.

15 **5.3.2 Characteristics of Sensitive Areas**

17 The ISA (U.S. EPA, 2008, Section 3.3)
 19 indicates that information is limited about the
 21 spatial extent and distribution of terrestrial
 23 ecosystems most sensitive to nutrient
 25 enrichment from atmospheric nitrogen
 27 deposition. Examples of sensitive ecosystems
 29 include the following:

“Effects are most likely to occur where areas of relatively high atmospheric nitrogen deposition intersect with nitrogen limited plant communities. The factors that govern the sensitivity...include the degree of nitrogen limitation, rates and form of atmospheric nitrogen deposition, elevation, species composition, length of growing season, and soil nitrogen retention capacity.”
 ISA, Section 3.3 (U.S. EPA 2008)

- 30 ▪ Alpine tundra (low rates of primary production, short growing season, low temperature,
 31 wide moisture variation, low nutrient supply).
- 32 ▪ Western United States ecosystems, such as the alpine ecosystems of the Colorado Front
 33 Range, chaparral watersheds of the Sierra Nevada Range, lichen communities in the San
 34 Bernardino Mountains and the Pacific Northwest, and CSS communities in Southern
 35 California.
- 36 ▪ Eastern United States ecosystems, where sensitivities are typically assessed in terms of the
 37 degree of nitrate leaching from soils into ground and surface waters. These ecosystems are

1 expected to include hardwood forests, semiarid lands and grassland ecosystems, but effects
2 on individual plant species have not been studied well.

3 In the Mediterranean systems of Southern California where rainfall is concentrated
4 during some months of the year, dry deposition is particularly important. Individual studies
5 measuring atmospheric nitrogen deposition to terrestrial ecosystems that involve throughfall
6 estimates for forested ecosystems can provide better approximations for total atmospheric
7 nitrogen deposition levels; however, such estimates and related bioassessment data are not
8 available for the entire country.

9 Finally, the exact relationship between atmospheric nitrogen loadings, fire frequency and
10 intensity, and nonnative plants, particularly in the CSS ecosystem, have not been quantified.
11 Various conceptual models linking these factors have been developed, but an understanding of
12 cause and effect, seasonal influences, and thresholds remains undeveloped.

13 The selection of case study areas specific to terrestrial nutrient enrichment began with
14 national GIS mapping to identify terrestrial areas potentially sensitive to atmospheric nitrogen
15 deposition. GIS datasets of physical, chemical, and biological properties that were indicative of
16 potential terrestrial nutrient enrichment were considered. The limited availability of datasets
17 resulted in selecting a combination of data on species sensitive or vulnerable to nitrogen
18 deposition combined with the exclusion of areas of the United States with anthropogenic
19 influence (e.g., urban, farmland).

20 Acidophytic lichens are known to be sensitive to increased levels of nitrogen loading. In
21 turn, other species are dependent upon lichens for both food and habitat. Locations where
22 acidophytic lichen were identified were defined as being sensitive.

23 Urban and agricultural land covers were also mapped, so they could be used to exclude
24 areas that are not sensitive to terrestrial nutrient enrichment, such as agricultural areas and
25 urbanized areas. Analysis of the presence of lichen over time compared to atmospheric nitrogen
26 deposition records and benchmarks can indicate the potential influence of nitrogen deposition.

27 Although there is no known nationwide species that has shown range loss because of
28 additional nitrogen, it was possible to assemble a “patchwork quilt” of species and forest types
29 from across the United States that are identified in the published literature as sensitive. These
30 species have evolved in settings across the United States to be able to assimilate specific levels
31 of nitrogen exposure. Some settings may naturally have low background concentrations of

1 nitrogen, so the species requires a relatively small amount to thrive. Other settings may have
2 higher background concentrations, where native species have evolved to thrive at those levels.
3 When exposed to nitrogen levels higher than natural background, the native species may be
4 vulnerable to invasion from species that use nitrogen in the shallow root zone before the nutrient
5 reaches deeper zones of the native ecosystem vegetation.

6 Soil nitrogen content data dating to pre-1980 were not available, and the quality of any
7 available data was uncertain. The physiographic provinces of the United States were considered
8 to provide leeward sides of mountains that tend to receive a greater amount of atmospheric
9 nitrogen deposition. However, this dataset was not used because terrain is already taken into
10 account by the CMAQ modeling.

11 The resulting map illustrates the areas of highest potential sensitivity (see **Figure 5.3-3**),
12 including coastal sage scrub, grasslands, and desert, as well as certain forest species and lichens.
13 This information facilitated the review of candidate case study areas.

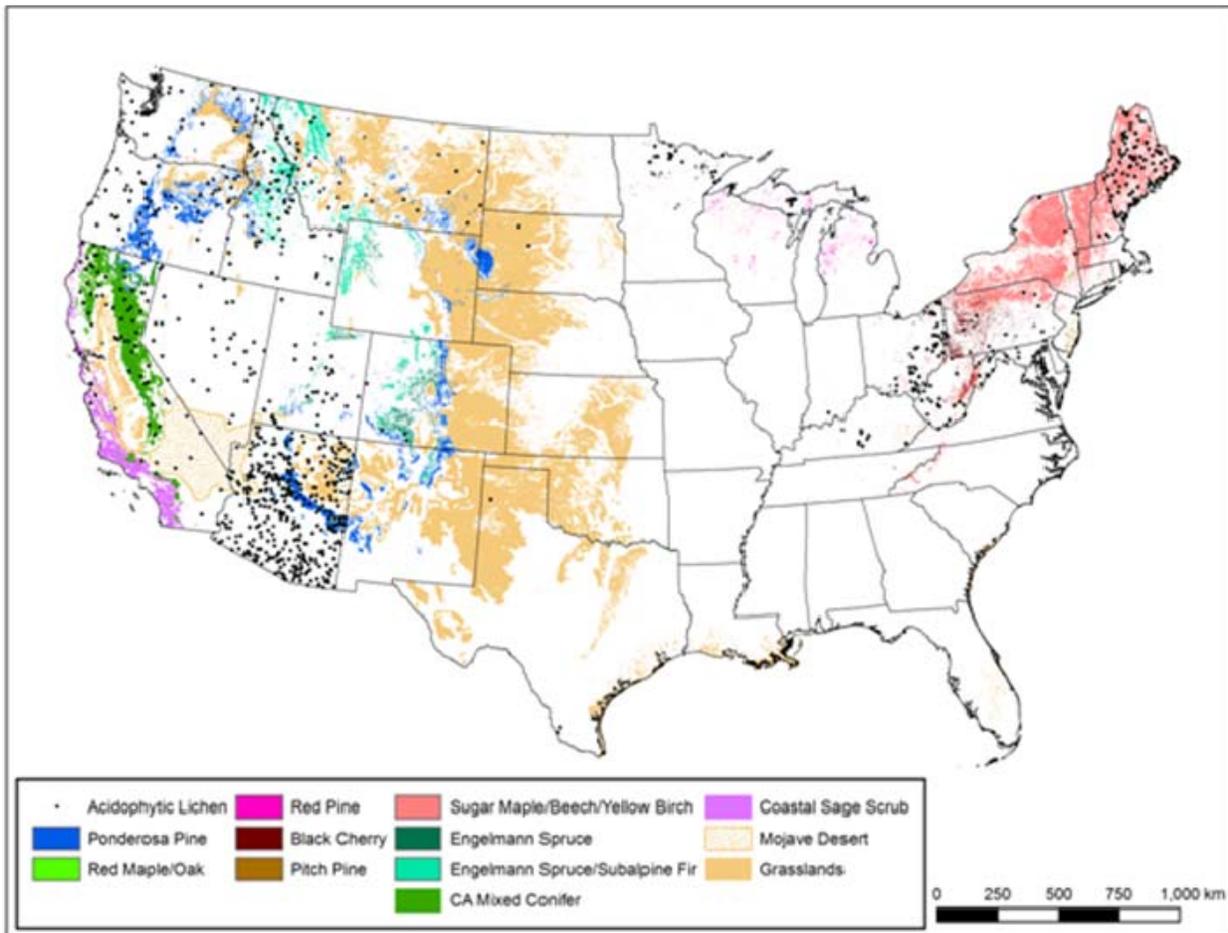
14 **5.3.3 Case Study Selection**

15 **Figure 5.3-3**, showing the areas of potential sensitivity to nutrient enrichment, was used
16 in conjunction with potential areas identified in the ISA (U.S. EPA, 2008, Section 4.3.1.2, Table
17 4.4) to select ecosystems for the case study. After considering this information, California's CSS
18 and MCF ecosystems were selected for this Terrestrial Nutrient Enrichment Case Study analysis
19 based on the following selection factors, in addition to the factors listed in Section 5.3.1:

- 20 ■ Availability of atmospheric ambient and deposition data (monitored or modeled)
- 21 ■ Availability of digitized datasets of biotic communities; fire-prone areas; and sensitive,
22 rare species
- 23 ■ Scientific results of research on nitrogen effects for the case study area
- 24 ■ Representation of western United States ecosystems potentially impacted by atmospheric
25 nitrogen deposition
- 26 ■ Scalability and generalization opportunities for risk analysis results from the case studies.

27 California's CSS has been the subject of intensive research in the past 10 years, which
28 has provided the data needed for a first phase of GIS analysis of the role of atmospheric nitrogen
29 deposition in terrestrial ecosystems. California's MCF has an even longer record of study that
30 includes investigations into the effects of atmospheric pollution, changes to forest structure,

1 changes to the lichen communities, and measurements of nitrogen saturation. Another ecosystem
 2 that was considered, but not selected for this case study, was the alpine ecosystem in the Rocky
 3 Mountains (see Section 5.3.6). As noted in the ISA (U.S. EPA, 2008, Section 3.3), results from a
 4 number of studies indicate that nitrates may be leaching from alpine catchments, and there
 5 appear to be changes in plant communities related to the deposition of atmospheric nitrogen. The
 6 amount of data from these alpine ecosystems is more limited than that from the CSS and MCF.
 7 However, the ecological benchmarks suggested for alpine ecosystems were comparable to the
 8 benchmarks from CSS and MCF ecosystems.



9
 10 **Figure 5.3-3.** Areas of highest potential nutrient enrichment sensitivity. (Acidophytic
 11 lichens, tree species, and the extent of the Mojave Desert come from data obtained from
 12 the United States Forest Service. The extents of coastal sage scrub and California mixed
 13 conifer forest come from the California Fire and Resource Assessment
 14 Program. Grasslands were obtained from the National Land Cover Dataset [USGS]).

5.3.4 Current Conditions in Case Study Areas

To assess current conditions, the ISA (U.S. EPA, 2008) provided the basis for identifying the published scientific literature on CSS and MCF ecosystems. In addition, spatially distributed data are available and support a GIS analysis. Section 2.2 of Appendix 7 describes the data sources used in the GIS analysis.

One of the central analytic tasks was to quantify the amount of CSS (and MCF loss) and to see whether this corresponded spatially to areas of high total nitrogen deposition or fire threat, or both.

CSS and MCF were selected as case study areas for the following reasons:

- Significant geographic coverage and they are located where urban areas interface with wilderness areas.
- Nitrogen deposition gradients, ranging from low background levels to some of the highest deposition levels recorded in the United States.
- Researched for extended periods to understand the interactive effects of deposition, climate change, fire, and other stressors.
- Research investigations are well documented in the peer-reviewed literature.

5.3.4.1 Coastal Sage Scrub

CSS is subject to several pressures, such as land conversion, grazing, fire, and pollution, all of which have been observed to induce declines in other ecosystems (Allen et al., 1998). At one extreme, development pressure (i.e., the conversion of CSS to residential and commercial land uses) will simply eliminate acres of CSS. Other pressures will come into play in modifying the remaining ecosystem. Research suggests that both fire and increased atmospheric nitrogen deposition can enhance the growth of nonnative grasses in established CSS communities. Additionally, CSS declines have been observed when fire frequency is held constant and/or nitrogen is held constant, suggesting that both fire and nitrogen play a role in CSS decline when direct destructive factors are not an imminent threat. Table 3.1-1 of Appendix 7 contains a summary of selected experimental variables across multiple CSS study areas.

Increased atmospheric nitrogen deposition has been observed to alter vegetation types when nitrogen is a limiting nutrient to growth. This is observed in alpine plant communities in the Colorado Front Range, as well as in lichen communities in the western Sierra Nevada region (Fenn et al., 2003, 2008); however, in the case of CSS, it is hypothesized that many stands are no longer limited by nitrogen and have instead become nitrogen-saturated because of atmospheric nitrogen deposition (Allen et al., 1998; Westman, 1981). This is supported by the positive correlation between atmospheric nitrogen and soil nitrogen, increased long-term mortality of

CSS shrubs, and increased nitrogen-cycling rates in soil and litter and soil fertility (Allen et al., 1998; Padgett et al., 1999; Sirulnik et al., 2007; Vourlitis et al., 2007). **Figure 5.3-4** illustrates the levels of atmospheric nitrogen deposition in CSS communities using CMAQ/NADP data.

Wood et al. (2006) 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 (Wood et al., 2006). It is assumed that 3.3 kg N/ha/yr is near the point where nitrogen is no longer limiting in the CSS community. Therefore, this amount can be considered an ecological benchmark for the CSS

community. **Figure 5.3-4** displays the spatial

extent of CSS where total nitrogen

deposition is above the ecological

benchmark of 3.3 kg N/ha/yr. **Table 5.3-1**

displays the areas (in hectares) of CSS

experiencing different total nitrogen deposition levels.

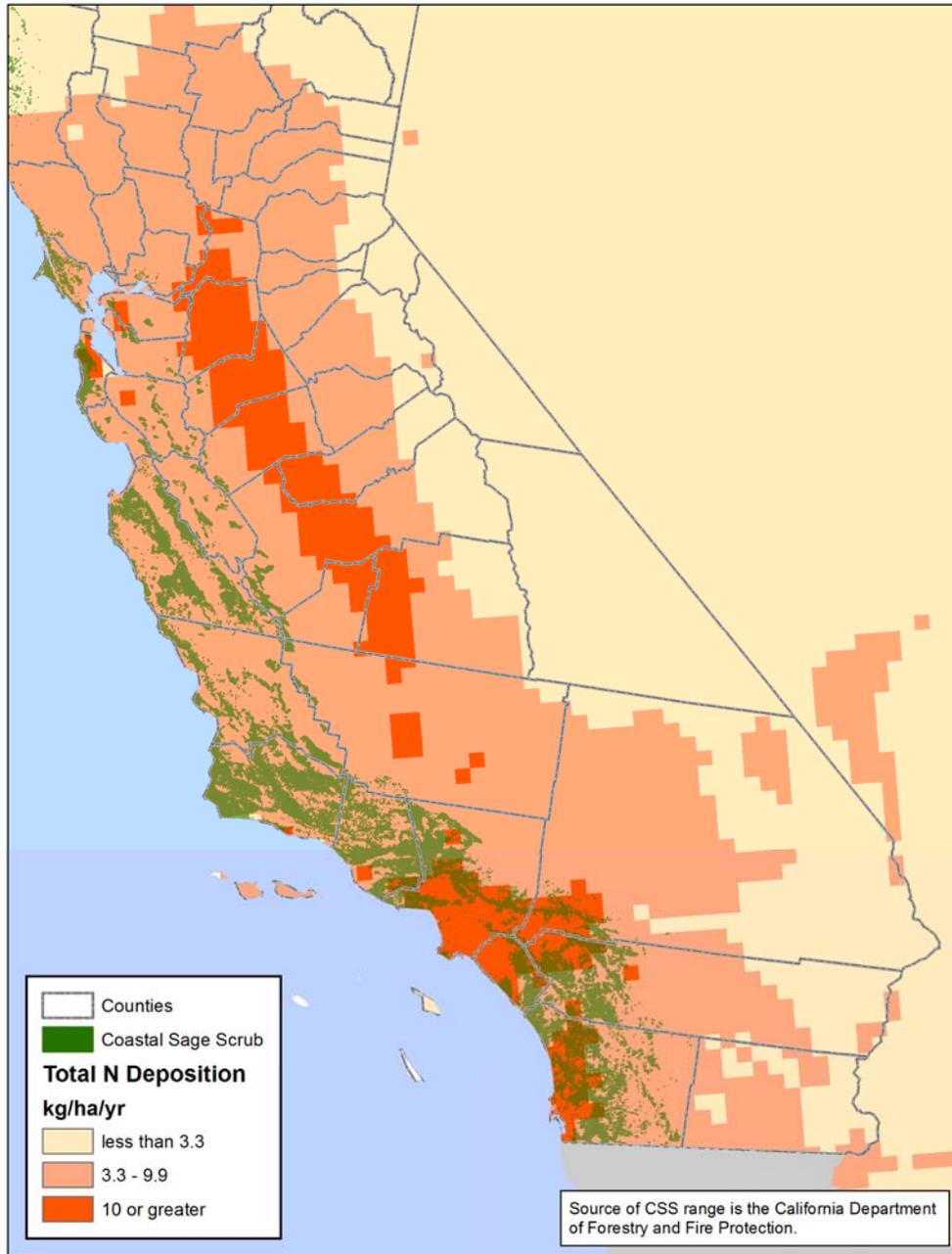
In the rainy, winter season, deposited surface nitrogen is transported deeper into the soil and is rapidly mineralized by microbes, favoring the germination and growth of nonnative grasses (e.g., *Bromus madritensis*, *Avena fatua*, and *Hirschfeldia incana*). Flourishing of grasses can create a dense network of shallow roots, which slows the diffusion of water through soil, decreases the percolation depth of precipitation, and decreases the amount of water for soil and ground water recharge (Wood et al., 2006). Growth of CSS species, such as *Artemisia californica*, *Eriogonum fasciculatum*, and *Encelia farinose*, may be reduced because of decreased water and nitrogen availability at the deeper soil layers where more woody CSS tap roots are found (Keeler-Wolf, 1995; Wood et al., 2006).

Mutualistic fungal communities, such as arbuscular mycorrhizae (AM) (Egerton-Warburton and Allen, 2000; Siguenza et al., 2006), increase the surface area and capacity for nutrient uptake. CSS is predominantly colonized by a coarse AM species, and nonnative grasses are more likely mutualistic with finer AM species. In the presence of elevated nitrogen, coarse AM colonizations were depressed in number and volume. Egerton-Warburton and Allen (2000) documented shifts in AM species as well as declines in spore abundance and colonization at approximately 10 kg N/ha/yr. Therefore, it is suggested that these reduced mutualistic associations of coarse AM may contribute to a decline in the overall health of CSS via a loss in

Table 5.3-1. Coastal Sage Scrub Ecosystem Area and Total Nitrogen Deposition

N Deposition (kg/ha/yr)	Area (hectares)	Percent of CSS Area, %
≥3.3	654048.4179	93.51
≥10	138019.8922	19.73

1 nutrient uptake capacity and represent an ecological benchmark for the CSS ecosystem. **Figure**
2 **5.3-4** displays the levels of total nitrogen deposition on CSS communities above the ecological
3 benchmark of 10 kg N/ha/yr using CMAQ/NADP data. The 12 km CMAQ/NADP data indicate
4 that CSS communities within the Los Angeles and San Diego airsheds are likely to experience
5 the noted effects at the 10 kg N/ha/yr ecological benchmark.



1

2

3

Figure 5.3-4. Coastal sage scrub range and total nitrogen deposition using CMAQ 2002 modeling results and NADP monitoring data.

4

5

6

7

8

Studies have suggested that plant-available nitrogen in soils may be increasing because of soil fertility in conjunction with atmospheric deposition, so that the soil itself becomes an intrinsic source of nitrogen (Padgett et al., 1999). In combination with decreased establishment and the capacity for nutrient uptake, these responses to elevated nitrogen levels may represent a detrimental and long-term pressure on CSS at varying levels of nitrogen additions. **Table 3.1-3**

1 of Appendix 7 summarizes the various ecosystem responses to nitrogen levels that affect CSS
2 communities.

3 Fire is also an inextricable and significant component in CSS losses. Although CSS
4 communities are fire resilient, nonnative grass seeds are quick to establish in burned lands,
5 reducing the water and nutrient amounts available to CSS for reestablishment (Keeler-Wolf,
6 1995). Additionally, when annual grasses have established dominance, these species alter and
7 increase the fire frequency as they senesce earlier in the annual season, which increases dry,
8 ignitable fuel availability (Keeley et al., 2005). With increased fire frequencies and faster
9 nonnative colonizations, CSS seed banks are eventually eradicated from the soil, and the
10 probability of reestablishment decreases significantly (Keeley et al., 2005). **Figure 5.3-5**
11 represents the fire threats to CSS communities.



1
2
3
4
5
6

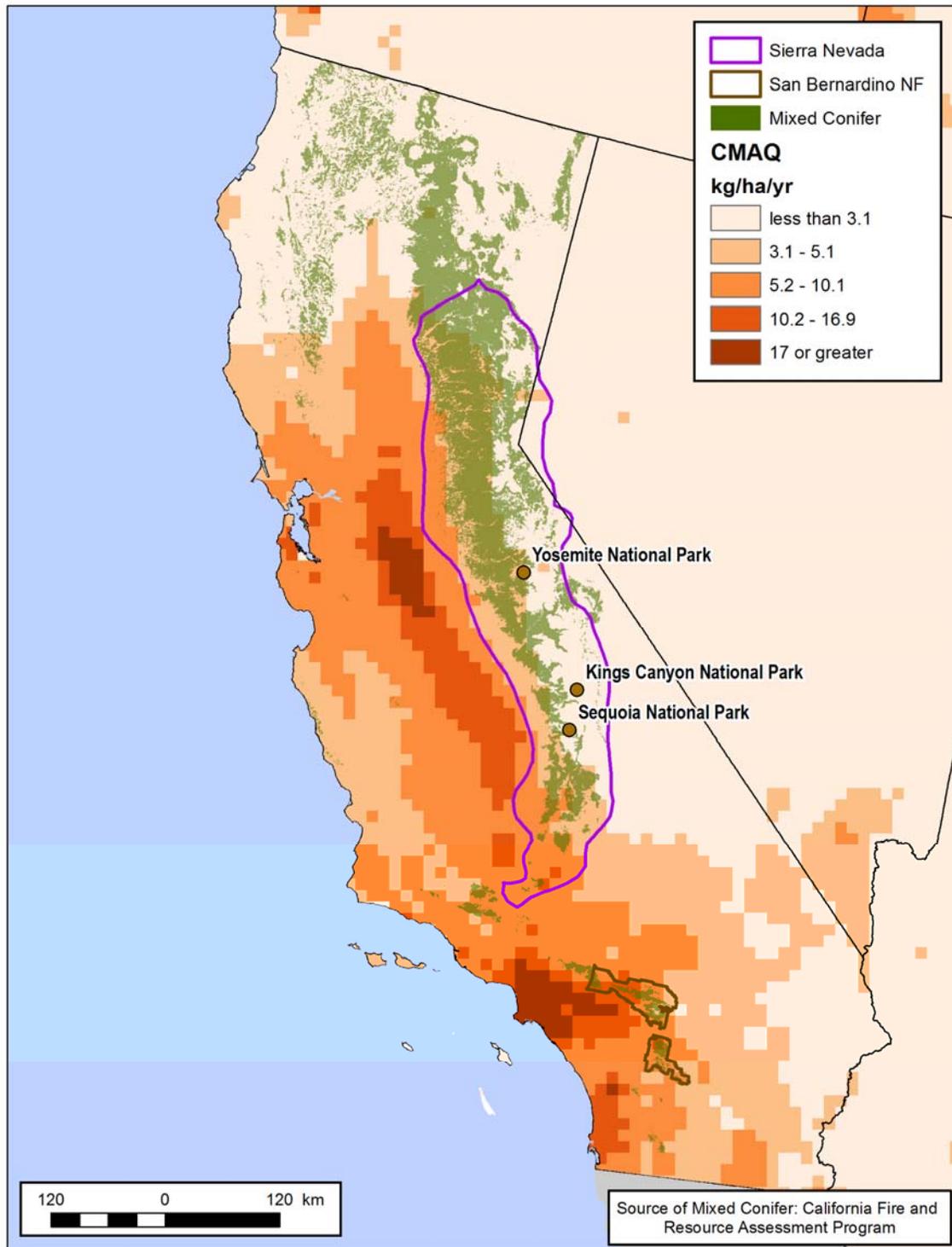
Figure 5.3-5. Current fire threats to coastal sage scrub communities.

5.3.4.2 Mixed Conifer Forest Ecosystems

The MCF ecosystem has been a subject of study for many years. There are a number of important stressors on the community, including fire, bark beetles, ozone, particulates, and atmospheric nitrogen. Although fire suppression in the 20th century is probably the most

1 significant change that has led to alterations in morphology and perhaps to shifts in forest
2 composition (Minnich et al., 1995), stress from elevated levels of ambient atmospheric nitrogen
3 concentrations is the subject of increasing research.

4 Measurements documenting increases in atmospheric nitrogen deposition have been
5 recorded with some regularity since the 1980s (Bytnerowicz and Fenn, 1996); however, the Los
6 Angeles area has seen elevated ambient atmospheric nitrogen concentrations for the last 50 years
7 (Bytnerowicz and Fenn, 1996). Also, some data have been published for the primary nitrogen
8 species of dry atmospheric nitrogen deposition in the San Bernardino Mountains (i.e., nitric acid
9 [HNO₃] and ammonia gas [NH₃]) from passive samplers (Bytnerowicz et al., 2007). The
10 pressures exerted on MCF ecosystems in California form a gradient across the Sierra Nevada
11 Range and San Bernardino Mountains. Nitrogen throughfall levels in the northern Sierra Nevada
12 Range are as low as 1.4 kg N/ha/yr, whereas forests in the western San Bernardino Mountains
13 experience measured throughfall nitrogen levels up to 33 to 71 kg N/ha/yr. (Note that the high
14 levels of nitrogen seen in some measured throughfall values are not reflected in the
15 CMAQ/NADP data.) The primary source of nitrogen in the western San Bernardino Mountains
16 stems from fossil fuels combustion, such as vehicle exhaust. Other sources, such as agricultural
17 processes, also play a prominent role in the western portions of the San Bernardino and Sierra
18 Nevada Range (Grulke et al., 2008). **Figure 5.3-6** illustrates the current total atmospheric
19 nitrogen deposition on MCF in California.



1
2
3

Figure 5.3-6. Mixed conifer forest range and total nitrogen deposition using CMAQ 2002 modeling results and NADP monitoring data.

1 At the individual tree level, elevated atmospheric nitrogen can shift the ratio of
2 aboveground to belowground biomass. Elevated pollution levels allow increased uptake of
3 nutrients via the canopy, reduced nitrogen intake requirements on root structures, and increased
4 demand for carbon dioxide (CO₂) uptake and photosynthetic structures to maintain the carbon
5 balances. Therefore, the increased nutrient availability stimulates aboveground growth and
6 increases foliar production while reducing the demand for belowground nutrient uptake (Fenn et
7 al., 2000) resulting in diminished fine-root biomass (Fenn and Bytnerowicz, 1997). Grulke et al.
8 (1998) observed a 6- to 14-fold increase in fine-root mass in areas of low atmospheric nitrogen
9 deposition as compared to areas of high deposition.

10 At the stand level, elevated atmospheric nitrogen has been associated with increased
11 stand density, although other factors, such as fire suppression and ozone, also contribute to
12 increased density and can increase mortality rates (U.S. EPA, 2008). As older trees die, they are
13 replaced with younger, smaller trees. Smaller trees allow more sunlight through the canopy and,
14 combined with an increased availability of nitrogen, may allow for more trees to be established.
15 Increased stand densities with younger-age classes are observed in the San Bernardino
16 Mountains, where air pollution levels are among the highest found in the California conifer
17 ranges studied (Minnich et al., 1995; Fenn et al., 2008). These shifts in stand density and age
18 distribution result in vegetation structure shifts which, in turn, may impact population and
19 community dynamics of understory plants and animals, including threatened and endangered
20 species.

21 It should be noted that the effects of ozone and atmospheric nitrogen are difficult to
22 separate. The atmospheric transformation of nitrogen oxides can yield moderate concentrations
23 of ozone as a byproduct (Grulke et al., 2008). Therefore, since elevated nitrogen levels are
24 generally correlated with ozone concentrations, researchers often report changes in tree health
25 and physiology as being the result of both (Grulke and Balduman, 1999).

26 High concentrations of ozone and atmospheric nitrogen can generate increased needle
27 and branch turnover. In areas subjected to low pollution, conifers may retain needles across 4 or
28 5 years; however, in areas of high pollution, such as Camp Paivika in the San Bernardino
29 Mountains, needle retention was generally less than 1 year (Grulke and Balduman, 1999; Grulke
30 et al, 2008). Needle turnover significantly increases litterfall. Litter biomass has been observed to
31 increase in areas with elevated atmospheric nitrogen deposition up to 15 times more than in areas

1 with low deposition (Fenn et al., 2000; Grulke et al., 2008). The increased litter deposition may
2 facilitate faster rates of microbial decomposition initially but may reduce decomposition over the
3 long term because of changes in the C:N ratio and increasing lignin content over time (Grulke et
4 al., 2008; U.S. EPA, 2008). The increased litter depth may then affect subcanopy growth and
5 stand regeneration over long periods of time.

6 At the highest levels of atmospheric nitrogen deposition, native understory species were
7 seen to decline (Allen et al., 2007). In addition to the decline in native understory diversity,
8 changes in decreased fine-root mass, increased needle turnover, and the associated
9 chemostructural alterations, MCF exposed to elevated pollutant levels have an increasing
10 susceptibility to drought and beetle attack (Grulke et al., 1998, 2001; Takemoto et al., 2001).
11 These stressors often result in the death of trees, producing an increased risk of wildfires.

12 Lichens emerged as an indicator of nutrient enrichment from the research on the effects
13 of acid rain. Lichen species are sensitive to air pollution; in particular, atmospheric nitrogen.
14 Since the 1980s, information about lichen communities has been gathered, and lichens have been
15 used as indicators to detect changes in forest communities.

16 As atmospheric nitrogen deposition increases, the relative abundance of acidophytic
17 lichens decreases, and the concentration of nitrogen in one of those species, *Letharia vulpine*,
18 increases (Fenn et al., 2008). Fenn et al. (2008) were able to quantify the change in the lichen
19 community, noting that for every 1 kg N/ha/yr increase, the abundance of acidophytic lichens
20 declined by 5.6%. **Figure 5.3-7** illustrates the presence of acidophyte lichens and the total
21 atmospheric nitrogen deposition in the California ranges.

22 In addition to abundance changes, species richness, cover, and health are affected in areas
23 of high ozone and nitrogen concentrations. Fifty percent fewer lichen species were observed after
24 60 years of elevated air pollution in San Bernardino Mountains MCF, with the areas of highest
25 pollution levels exhibiting low species richness, decreased abundance and cover, and
26 morphological deterioration of existing lichens (Sigal and Nash, 1983).

27 Fenn et al. (2008) found that at 3.1 kg N/ha/yr, the community of lichens begins to
28 change from acidophytic to tolerant species; at 5.2 kg N/ha/yr, the typical dominance by
29 acidophytic species no longer occurs; and at 10.2 kg N/ha/yr, acidophytic lichens are totally lost
30 from the community. Additional studies in the Colorado Front Range of the Rocky Mountain
31 National Park support these findings and are summarized in Chapter 5.3.6.2 of this Risk and

1 Exposure Assessment. These three values (3.1, 5.2, and 10.2 kilograms per hectare per year
2 [kg/ha/yr]) are one set of ecologically meaningful benchmarks for the MCF. Figure 5.3-7 shows
3 the presence of acidophytic lichen species above the three ecological benchmarks. Nearly all of
4 the MCF communities receive total nitrogen deposition levels above the 3.1 N kg/ha/yr
5 ecological benchmark according to the 12- km 2002 CMAQ/NADP data, with the exception of
6 the easternmost Sierra Nevada Range. MCF in the southern portion of the Sierra Nevada forests
7 and nearly all MCF communities in the San Bernardino forests receive total nitrogen deposition
8 levels above the 5.2 N kg/ha/yr ecological benchmark. **Figure 5.3-7** also displays the potential
9 areas where acidophytic lichens are extirpated because of nitrogen deposition levels above 10.2
10 kg N kg/ha/yr.

11 The established signs of nitrogen saturation have been shown within the MCF ecosystem.
12 These symptoms include the following:

- 13 ■ Increased carbon and nitrogen cycling
- 14 ■ Decreased nitrogen uptake efficiency of plants
- 15 ■ Increased loss of forest nitrates to streamwater (NO_3^- leachate).

16 Fenn et al. (2008) established a critical loading benchmark of 17 kg throughfall N/ha/yr
17 (which is the actual nitrogen deposited on the forest floor as opposed to modeled nitrogen
18 deposition) in the San Bernardino and Sierra Nevada Range MCF ecosystems. This benchmark
19 represents the level of atmospheric nitrogen deposition at which elevated concentrations of
20 streamwater NO_3^- leachate or potential nitrogen saturation may occur. At this deposition level, a
21 26% reduction in fine-root biomass is anticipated (Fenn et al., 2008). Root:shoot ratios are,
22 therefore, altered, and changes in nitrogen uptake efficiencies, litterfall biomass, and microbial
23 decomposition are anticipated to be present at this atmospheric nitrogen deposition level. This
24 benchmark is based on 30 to 60 years of exposure to elevated atmospheric concentrations. At
25 longer exposure levels, the benchmark is lower because of decreased nitrogen efficiencies of the
26 ecosystem. This benchmark is exceeded in areas of the western San Bernardino Mountains, such
27 as Camp Paivika.

28 Nitrate leaching is a symptom that an ecosystem is saturated by nitrogen. Nitrate leaching
29 is also known to cause acidification in adjacent surface waters. The ecological benchmark of 17
30 kg N/ha/yr is the last benchmark identified in this study. At this level of atmospheric nitrogen

1 deposition, nitrate is observed in streams in the MCF (Fenn et al., 2008), denoting a change in
 2 ecosystem function.

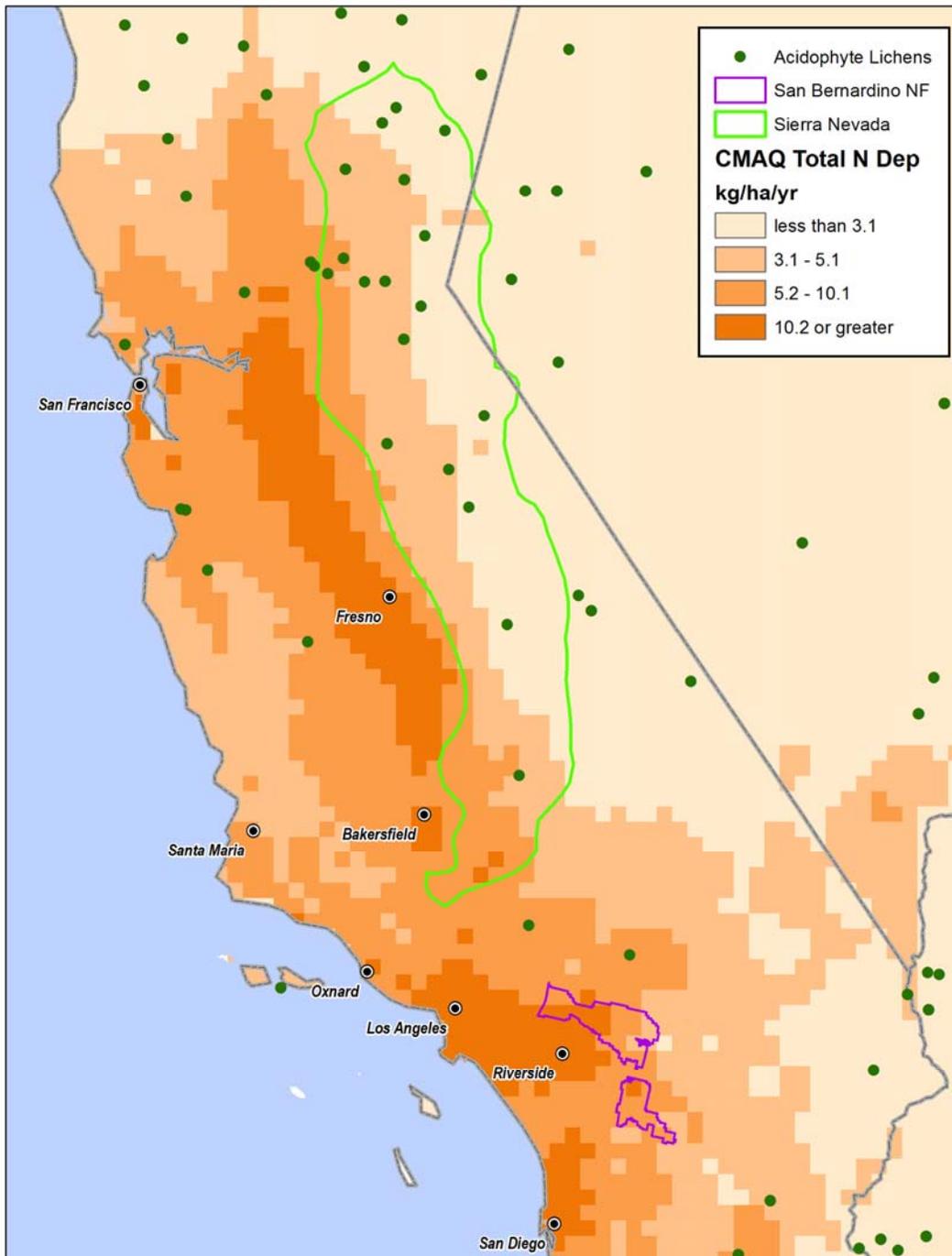
3 **Table 5.3-2** shows the area of MCF experiencing levels of nitrogen deposition
 4 corresponding to the identified benchmarks.

5 **Table 5.3-2.** Mixed Conifer Forest Ecosystem Area and Nitrogen Deposition

N Deposition (kg/ha/yr)	Area (hectares)	Percent of MCF Area, %
≥3.1	1099133.482	38.62
≥5.2	130538.2573	4.59
≥10.2	11963.08815	0.42
≥17	0	0.00

6 **Note:** According to the 12-km CMAQ data, there is too little
 7 area receiving >17 kg N/ha/yr to be measurable.

8



1
2 **Figure 5.3-7.** Presence of acidophyte lichens and total nitrogen deposition in the California
3 mountain ranges using CMAQ 2002 modeling results and NADP monitoring data.

4 **5.3.5 Degree of Extrapolation to Larger Assessment Areas**

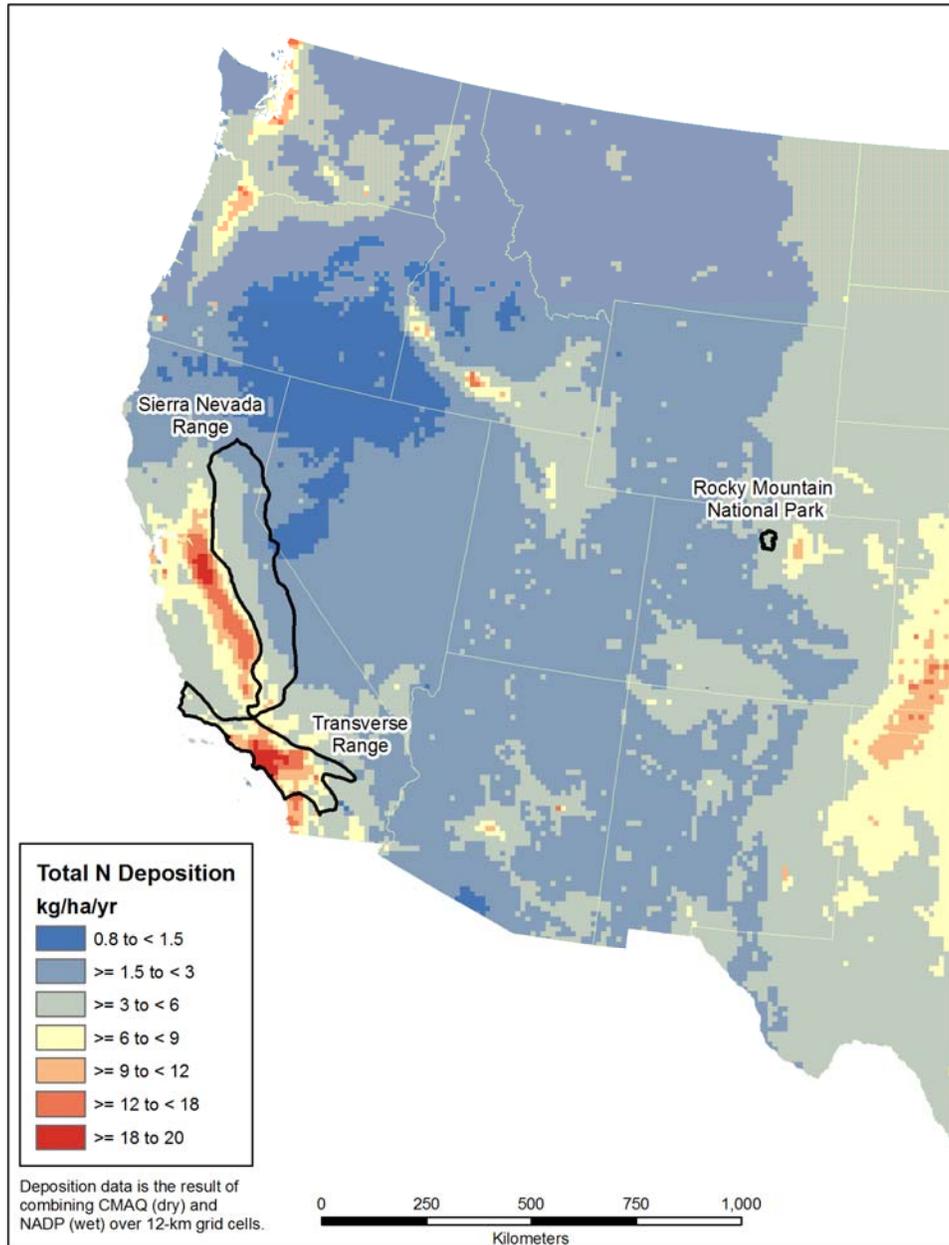
5 The Terrestrial Nutrient Enrichment Case Study examined the effects of atmospheric
6 nitrogen on two ecosystem types in California, CSS and MCF. **Figure 5.3-8** presents CMAQ

1 2002–modeled and NADP-monitored deposition of total nitrogen in the western United States. In
2 the western United States, other arid and forested ecosystems exposed to deposition at levels
3 discussed in this case study may experience altered effects. As noted in the previous section,
4 research on grasslands and chaparral ecosystems is underway. Nitrate leaching in forests with
5 elevated deposition may result in nitrate leaching that subsequently enriches and affects aquatic
6 ecosystems. Research on lichen species in the Pacific Northwest and in Central California that
7 are also exposed to elevated levels of atmospheric nitrogen deposition is also being conducted.
8 Extensive research on the eastern Front Range of the Rocky Mountain National Park has been
9 conducted in alpine and subalpine terrestrial and aquatic systems at elevations about 3,300
10 meters (m), where communities are typically adapted to low nutrient availability but are now
11 being exposed to >10 kg N/ha/yr in some study areas.

12 Locations were identified where data were available that might have implications for
13 other ecosystems and ecosystem services, as well as where a compelling case may be found to
14 show that the effects were due to atmospheric deposition of nitrogen. Other systems that are also
15 sensitive might include the following:

- 16 ■ **Ecosystems with nitrogen-sensitive epiphytes, such as lichens or mycorrhizae.** Such
17 systems may demonstrate shifts in community structure through changes in nutrient
18 availability or modified provisioning services.
- 19 ■ **Ecosystems that may have been exposed to long periods of elevated atmospheric**
20 **nitrogen deposition.** The established signs of nitrogen saturation are increased leaching of
21 NO_3^- into streamwater, decreased nitrogen uptake efficiency of plants, and increased
22 carbon and nitrogen cycling. At prolonged elevated nitrogen levels, ecosystems are
23 generally less likely to use, retain, or recycle nitrogen species efficiently at both the
24 species and community levels.
- 25 ■ **Critical habitats.** Ecosystems that are necessary for endemic species or special ecosystem
26 services should be monitored for possible changes due to nitrogen.
- 27 ■ **Locations where there are seasonal releases of nitrogen.** In both the California CSS and
28 MCF ecosystems discussed in the case study, a large portion of nitrogen is dry-deposited
29 and remains on the foliage and soil surface until the beginning of the winter rainy season
30 when nitrogen will be flushed into the soil.

1 In addition to the classic signs of nitrogen saturation, it is interesting to note that both
 2 CSS and the MCF ecosystems had responses in epiphytic associations, as well as increased
 3 susceptibility to wildfire and invasion. Water use was also modified in these systems. The
 4 implication and inferential magnitude of these results may warrant future investigations.



5
 6 **Figure 5.3-8.** CMAQ 2002 modeling results and NADP monitoring data for
 7 deposition of total nitrogen in the western United States.

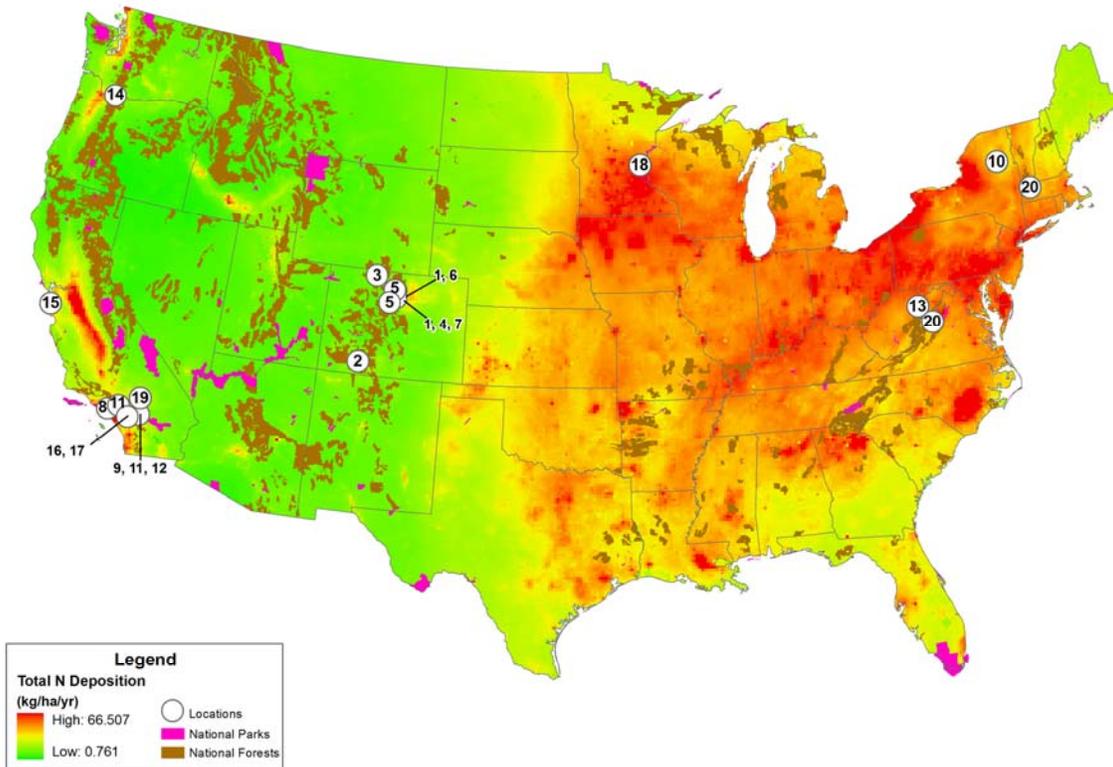
5.3.6 Current Conditions for Select Locations Nationwide

5.3.6.1 Overview

Figure 5.3-9 displays a map of observed effects from ambient and experimental atmospheric nitrogen deposition loads in relation to CMAQ 2002 modeling results and NADP monitoring data. The map depicts the sites where empirical effects of terrestrial nutrient enrichment have been observed and site proximity to elevated levels of atmospheric nitrogen deposition. The ISA (U.S. EPA, 2008, Section 3.3) also identifies areas of the western United States where atmospheric nitrogen deposition effects have been reported.

A range of ecological benchmarks were developed in the results. All benchmarks are tied to a level of atmospheric nitrogen deposition but include a number of different ecological processes. All of the benchmarks are ecologically significant in that changes that are related to community structure and function are seen. The benchmarks span a range from 3.1 to 17 kg N/ha/yr (see **Figure 5.3-2**) and include the following:

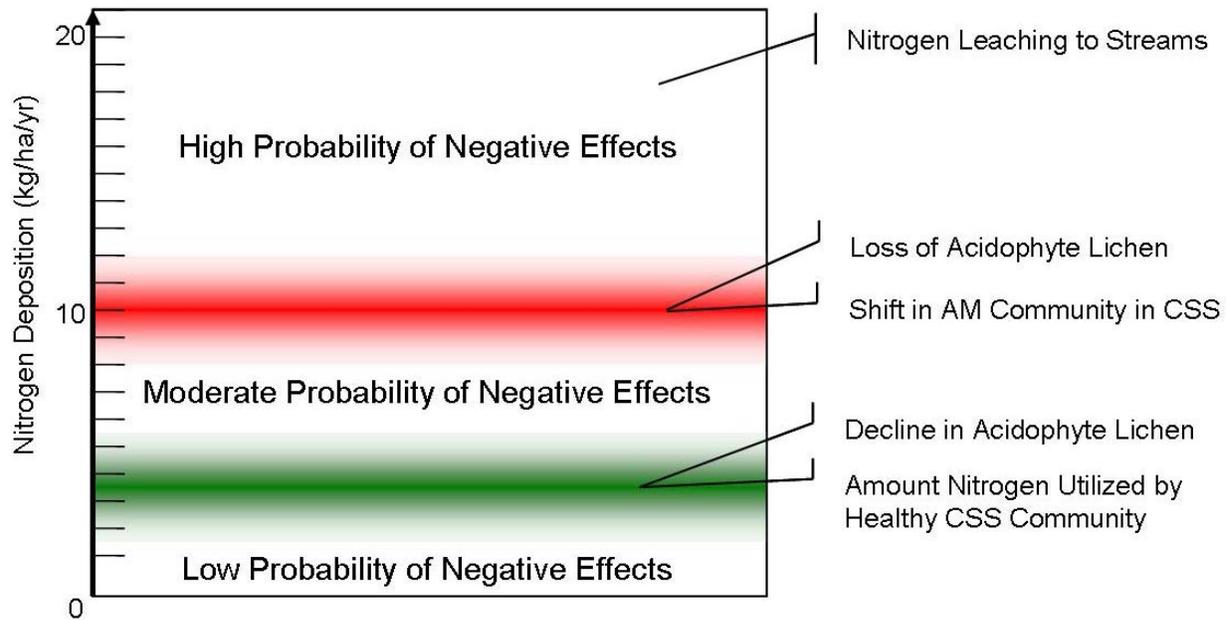
- 3.1 kg N/ha/yr – shift from sensitive to tolerant lichen species in MCF
- 3.3 kg N/ha/yr – the amount of nitrogen uptake by a vigorous stand of CSS; above this level, nitrogen may no longer be limiting
- 5.2 kg N/ha/yr – dominance of tolerant lichen species in MCF
- 10 kg N/ha/yr – mycorrhizal community changes in CSS
- 10.2 kg N/ha/yr – loss of sensitive lichen species in MCF
- 17 kg N/ha/yr – nitrate leaching into streams in MCF.



1. Nitrogen enrichment or eutrophication of lakes (Loch Vale, CO: 0.5 to 1.5 kg/ha/yr; Niwot Ridge, CO: 4.71 kg/ha/yr)
2. Alpine lakes increase shift in diatom species (Rocky Mountains, CO: 2 kg/ha/yr)
3. Alpine meadows' elevated NO_3^- levels in runoff (Colorado Front Range: 20, 40, 60 kg/ha/yr)
4. Alpine meadows' shift toward hairgrass (Niwot Ridge, CO: 25 kg/ha/yr)
5. Nitrogen enrichment or nitrogen saturation (e.g., soil and foliar nitrogen concentration) (eastern slope of Rocky Mountains: 1.2, 3.6 kg/ha/yr; Fraser Forest, CO: 3.2 to 5.5 kg/ha/yr)
6. Increased nitrogen mineralization rates and nitrification (Loch Vale, CO (spruce): 1.7 kg/ha/yr)
7. Alpine tundra with increased plant foliage and reduced species richness (Niwot Ridge, CO: 50 kg/ha/yr)
8. Nitrogen saturation, high NO_3^- in streamwater, soil, leaves; high nitric oxide (NO) emissions (Los Angeles, CA, air basin: saturation at 24 to 25 kg/ha/yr (dry) and at 0.8 to 45 kg/ha/yr (wet); northeastern U.S.: 3.3 to 12.7 kg/ha/yr)
9. Nitrogen saturation, high NO_3^- in streamwater (San Bernardino Mountains, CA (coniferous): 2.9 and 18.8 kg/ha/yr)
10. NO_3^- leaching (New England; Adirondack lakes: 8 to 10 kg/ha/yr)
11. Nitrogen saturation, high dissolved inorganic nitrogen (San Bernardino Mountains, San Gabriel Mountains, CA, chaparral, hardwood, coniferous): 11 to 40 kg/ha/yr)
12. Increased tree mortality and beetle activity (San Bernardino Mountains, CA (Ponderosa): 8 and 82 kg/ha/yr)
13. Enhanced growth of black cherry and yellow poplar; possible decline in red maple vigor; increased foliar nitrogen (Fernow Forest, WV: 35.5 kg/ha/yr)
14. Impacts on lichen communities (California MCF: 3.1 kg/ha/yr; Columbia R. Gorge, OR/WA: 11/5 to 25.4)
15. Evidence that threatened and endangered species impacted San Francisco Bay, CA (checkerspot butterfly and serpentine grass invasion: 10 to 15 kg/ha/yr; Jasper Ridge, CA: 70 kg/ha/yr)
16. Decreased diversity of mycorrhizal communities (Southern California: ~10 kg/ha/yr)
17. Decreased abundance of CSS (Southern California: 3.3 kg/ha/yr)
18. Loss of grasslands (Cedar Creek, MN: 5.3 [1.3 to 9.8] kg/ha/yr)
19. Decrease in abundance of desert creosote bush, increase in nonnative grasses (Mojave Desert and Chihuahuan Desert, CA: 1.7 kg/ha/yr and up)
20. Decrease in pitcher plant population growth rate (Hawley Bog, MA and Molly Bog, VA: 10 to 14 kg/ha/yr)

Figure 5.3-9. Observed effects from ambient and experimental atmospheric nitrogen deposition loads in relation to using CMAQ 2002 modeling results and NADP monitoring data. Citations for effect results are from the ISA, Table 4.4 (U.S. EPA, 2008)

1 This range of ecological benchmarks may be used to develop a “green line/red line”
 2 schematic, similar to the forest screening model discussed in Lovett and Tear (2007) that
 3 illustrates the levels at which ecosystem effects may occur or are known to occur. In **Figure**
 4 **5.3-10**, the green area/line denotes that point at which there do not appear to be any effects, and
 5 the red line denotes the point at which known negative effects occur.

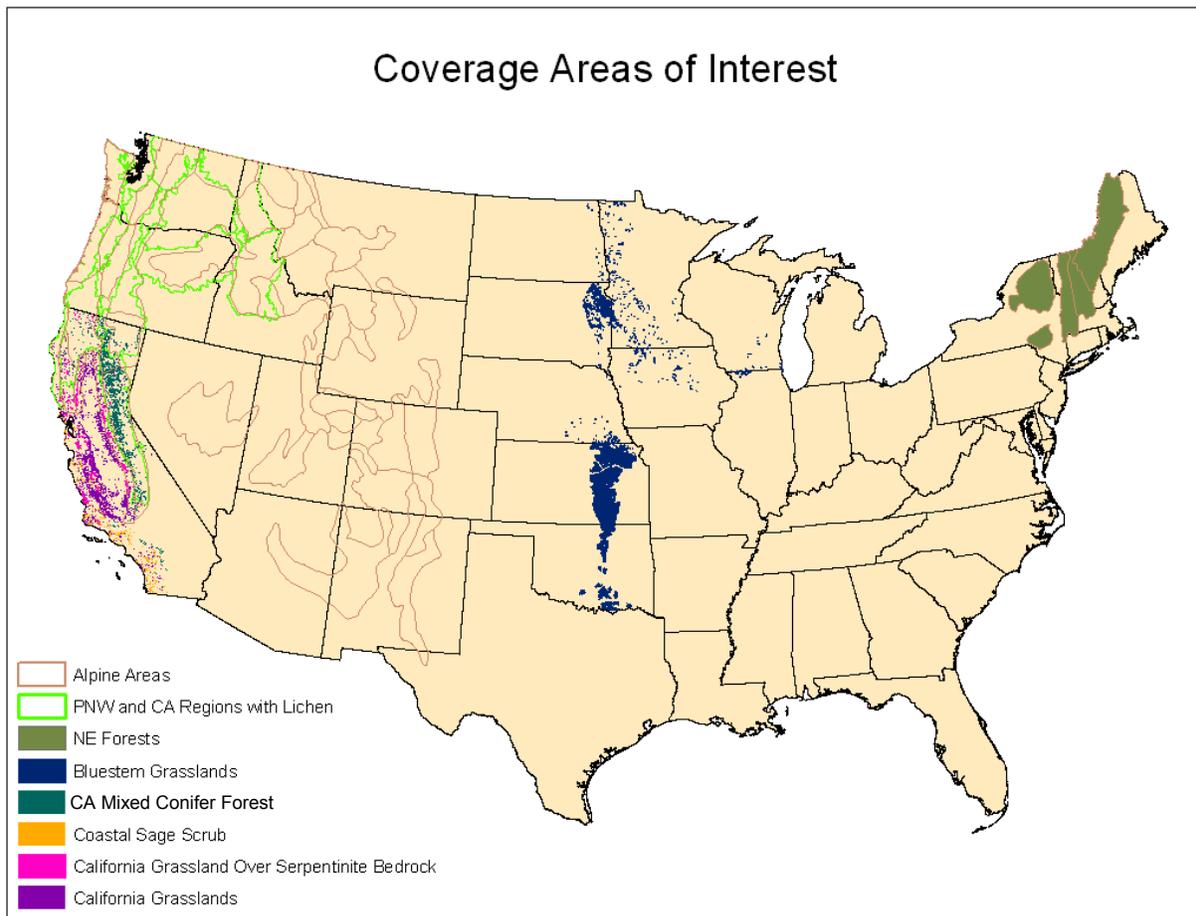


6
 7 **Figure 5.3-10.** Illustration of the range of terrestrial ecosystem effects observed
 8 relative to atmospheric nitrogen deposition.

9 For the benchmarks identified, effects may occur at the level of atmospheric nitrogen
 10 deposition associated with the “green line” illustrated in **Figure 5.3-10**, so the “green line” may
 11 be somewhat lower. The higher levels of atmospheric nitrogen deposition (both at 10.2 and 17
 12 kg/ha/yr) better resemble a “red line,” where a known negative effect occurs.

13 The range of ecological benchmarks in CSS and MCF are not dissimilar from those
 14 identified in other ecosystems with related characteristics, such as arid systems, other forested
 15 systems, or grasslands (see **Figure 5.3-11**). Egerton-Warburton et al. (2001) report that at 10 kg
 16 N/ha/yr, nitrogen changes in mycorrhizal communities/grass biomass are observed in chaparral
 17 ecosystems. Nitrates are found to leach into streams from nitrogen-saturated forest soils at
 18 deposition levels between 9 and 13 kg N/ha/yr (Aber et al., 2003). Results from several studies
 19 suggest ecosystem changes that are related to atmospheric nitrogen deposition. The capacity of
 20 alpine catchments to sequester nitrogen is exceeded at input levels <10 kg N/ha/yr (Baron et al.,

1 1994). Changes in the *Carex* plant community were observed to occur at deposition levels near
 2 10 kg N/ha/yr (Bowman et al., 2006). Clark and Tilman (2008) predict that at 5.3 kg N/ha/yr,
 3 there is a loss of species diversity in grasslands. In the Pacific Northwest and in Central
 4 California, a number of investigators have observed declines in sensitive lichen species as air
 5 pollution increases (Jovan and McCune, 2005; Geiser and Neitlich, 2007). In Europe, acidophyte
 6 decline has been identified in regions with 8 to 10 kg N/ha/yr (Bobbink, 1998; Bobbink et al.,
 7 1998).



8

9

10

Figure 5.3-11. Habitats that may experience ecological benchmarks similar to coastal sage scrub and mixed conifer forest.

11

5.3.6.2 Atmospheric Nitrogen Deposition Influence on Eastern Slope of Rocky Mountains

12

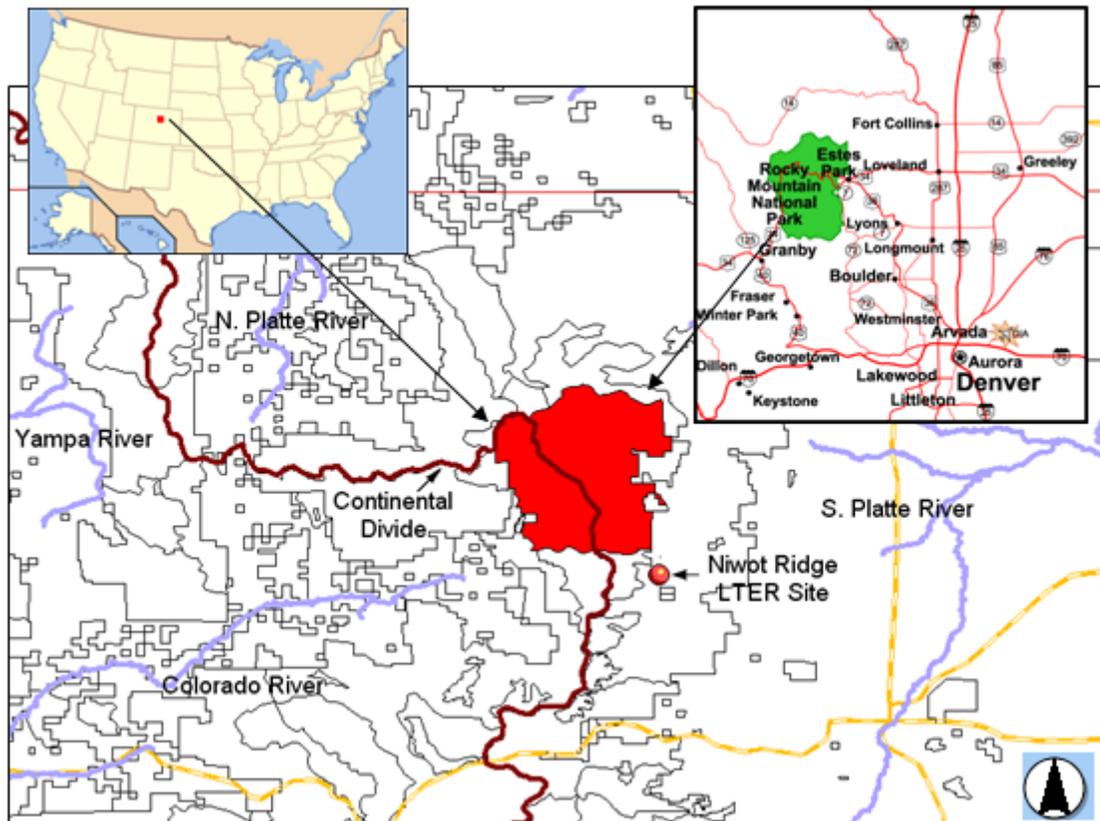
13

14

Rocky Mountain National Park encompasses approximately 265,770 acres (1,076 km²) of land in Colorado's northern Front Range. The park is split by the Continental Divide, which

1 gives the eastern and western portions of the park a different character. The east side of the park
2 tends to be drier, with heavily glaciated peaks and cirques. The west side of the park is wetter
3 and lush, dominated by deep forests. The park contains 150 lakes and 450 miles (720 km) of
4 streams. The park contains more than 60 named peaks higher than 12,000 feet (3,700 m), and
5 over one-fourth of the park resides above tree line. The lowest elevations in the park are montane
6 forests and grassland. The ponderosa pine, which prefers drier areas, dominates, though at higher
7 elevations Douglas fir trees are found. Above 9,000 feet (2,700 m) the montane forests give way
8 to the subalpine forest. Engelmann spruce and subalpine fir trees are common in this zone. These
9 forests tend to have more moisture than the montane and tend to be denser. Above tree line, at
10 approximately 11,500 feet (3,500 m), trees disappear, and it becomes alpine tundra.

11 Since Rocky Mountain National Park spans the Continental Divide, there are higher
12 levels of atmospheric nitrogen deposition to the east (the Front Range) than for the western parts
13 of the park due to transport of emissions from densely populated areas (e.g., the Denver
14 metropolitan area). Most of the detailed scientific studies documenting acid rain effects have
15 involved alpine or subalpine settings, usually at elevations of 3,100 m or more above mean sea
16 level. Rocky Mountain National Park is surrounded by other federal public lands. The Niwot
17 Ridge Long-Term Ecological Research (LTER) site is located in the Roosevelt National Forest
18 to the immediate southwest of Rocky Mountain National Park, and Niwot Ridge research
19 findings have applicability to patterns relevant to the Front Range (west of the Continental
20 Divide) portions of Rocky Mountain National Park. **(Figure 5.3-12)**



1
2 **Figure 5.3-12.** Rocky Mountain National Park location relative to the Niwot Ridge Long-
3 Term Ecological Research site and Denver metropolitan area.

4 **Aquatic Systems: Lakes and Streams.** Some alpine lakes in the west (including the
5 Rocky Mountains) show a seasonal pattern of episodic acidification for lakes (and also for
6 streams) from melting of snowpack in the early spring, related to poor acid neutralizing capacity
7 of the sparse soils and receiving waters and flushing of dissolved organic carbon (Denning et al.,
8 1991; Williams and Tonnessen, 2000). The hydrologic cycle in higher elevation areas is
9 dominated by the annual accumulation and melting of a dilute, mildly acidic snowpack. While
10 these areas are not as sensitive as other parts of the West, the ISA (U.S. EPA, 2008) presents
11 information showing that lakes in the Rocky Mountain area have been documented as acid-
12 sensitive waters in the EPA Western Lakes Survey (Landers et al., 1987; Stoddard et al., 2003).
13 Chronic acidification effects (e.g., as in the Adirondacks, are not prevalent for western lakes, but
14 episodic acidification has been reported for lakes in the Colorado Front Range [Brooks et al.,
15 1996; Williams et al., 1996]).

1 The ISA (U.S. EPA, 2008) presents scientific studies that show that increased
2 atmospheric nitrogen deposition in lakes and streams can cause a shift in community
3 composition and reduce algal biodiversity. Elevated nitrogen deposition results in changes in
4 algal species composition, especially in sensitive oligotrophic lakes. Field experiments show
5 responses to nitrogen for two opportunistic diatom species, *Asterionella formosa* and *Fragilaria*
6 *crotonensis* (McKnight et al., 1990; Lafrancois et al., 2004; Saros, 2005). These species now
7 dominate the flora of at least several alpine and montane Rocky Mountain lakes, with similar
8 field data showing shifts in dominant algal species in other parts of the West. These shifts in the
9 dominant algal species show up in Front Range lakes starting in the 1950s (Baron, 2006; Das et
10 al., 2005; Enders et al., 2008; Wolfe et al., 2001, 2003). Ambient nitrogen levels associated with
11 maximum species diversity for alpine lakes are estimated to be at or <3 micromoles (μM) based
12 on studies in the Yellowstone National Park (Interlandi and Kilham, 1998). A hindcasting
13 exercise has concluded that the change in Rocky Mountain National Park lake algae that
14 occurred between 1850 and 1964 was associated with an increase in wet nitrogen deposition that
15 was only about 1.5 kg N/ha (Baron, 2006). Similar changes inferred from lake sediment cores of
16 the Beartooth Mountains of Wyoming also occurred at about 1.5 kg N/ha deposition (Saros et al.,
17 2003).

18 **Terrestrial Systems.** Because alpine plant species are typically adapted to low nutrient
19 availability, they often are sensitive to effects from nutrient enrichment. The ISA (U.S. EPA,
20 2008) presents results from several studies suggesting that the capacity of Rocky Mountain
21 alpine catchments to sequester nitrogen is exceeded at input levels of about 4 kg N/ha/yr (Baron
22 et al., 1994; Williams and Tonnessen, 2000). For the Front Range, atmospheric deposition levels
23 are typically 3 to 5 kg N/ha/yr, with nitrogen deposition levels of 1 to 2 kg N/ha/yr typical in the
24 areas to the west of the Continental Divide (Baron et al., 2000).

25 Research on nutrient enrichment effects on alpine and subalpine ecosystems in the
26 western U.S. has been limited mainly to studies at the Loch Vale Watershed in Rocky Mountain
27 National Park and the Niwot Ridge LTER site, both located east of the Continental Divide in
28 Colorado (Burns, 2004). Research has been conducted in this area on both the terrestrial and
29 aquatic effects of nutrient enrichment. At these locations, experiments have involved controlled
30 fertilization to document the effects on species composition simulating the effects of nitrogen

1 atmospheric deposition. Increased cover and total biomass of both grasses and sedges (*Carex*
2 spp.) was a common response pattern.

3 High elevation alpine terrestrial communities exhibit a relatively low capacity to
4 sequester atmospheric deposition of nitrogen because of steep slopes, shallow soils, sparse
5 vegetation, short growing season, and other factors (Baron et al., 1994; Williams et al., 1996).
6 Results from several studies suggest that the capacity of individual indicator species in Rocky
7 Mountain alpine catchments to sequester nitrogen is exceeded at deposition levels of 3-4 kg
8 N/ha/yr (Baron et al., 1994; Williams and Tonnessen, 2000). Effects of N_r deposition to alpine
9 terrestrial ecosystems in this area could include community-level changes in plants, lichens, and
10 mycorrhizae. A variety of species could serve as useful indicators. The changes in plant species
11 that occur in response to atmospheric nitrogen deposition in the alpine zone can result in further
12 increased leaching of NO_3^- from the soils, because the plant species favored by higher nitrogen
13 supply are often associated with greater rates of nitrogen mineralization and nitrification than the
14 preexisting species (Bowman et al., 1993, 2006; Steltzer and Bowman, 1998; Suding et al.,
15 2006).

16 The ISA (U.S. EPA, 2008) presents results from several studies that suggest the capacity
17 of Rocky Mountain alpine catchments to sequester nitrogen is exceeded at input levels <3-4 kg
18 N/ha/yr (Baron et al., 1994; Williams et al., 1996). Changes in an individual species (*Carex*
19 *rupestris* and *Trisetum spicatum*) were estimated to occur at deposition levels near 4 kg N/ha/yr
20 (Bowman et al., 2006). Changes in the community, based on the first axis of a detrended
21 correspondence analysis, were estimated to occur at deposition levels near 10 kg N/ha/yr.
22 (Bowman et al., 2006). In comparison, critical loads for alpine plant communities in Europe are
23 5 to 15 kg N/ha/yr (Bobbink, 1998). It is also worth noting that some state agencies have pursued
24 the use of critical loads independently to link science and policy in addressing the management
25 of natural resources. For instance, in the State of Colorado, critical loads for atmospheric
26 nitrogen deposition that were developed for Rocky Mountain National Park (Baron, 2006) are
27 being used to develop goals for nitrogen emissions reductions by the State of Colorado, U.S.
28 EPA, and NPS. (See “Nitrogen Deposition Reduction Plan” at
29 <http://www.cdphe.state.co.us/ap/rmnp.html>)

30 Effects of N_r deposition to alpine terrestrial ecosystems in this area could include
31 community-level changes in plants, lichens, and mycorrhizae. A variety of species could serve as

1 useful indicators. The ISA (U.S. EPA, 2008) notes that there are difficulties, however, in
2 correlating community or indicator species responses exclusively with atmospheric nitrogen
3 deposition. In many instances, the confounding influences of climatic change, particularly
4 changes in precipitation, cannot be ruled out (Williams et al., 1996; Sherrod and Seastedt, 2001;
5 Fenn et al., 2003).

6 **5.3.7 Ecological Effect Function for Terrestrial Nutrient Enrichment**

7 There are many factors that determine whether or not an ecological effect occurs in
8 response to ambient concentrations of NO_x and SO_x. These may be ecological or atmospheric
9 factors, both of which influence deposition or exposure and the subsequent ecological effects
10 (i.e., acidification or nutrient enrichment). In the Terrestrial Nutrient Enrichment Case Study,
11 establishing a quantitative linkage between a given ecological indicator and deposition, as
12 influenced by the variable ecological factors, was not addressed because deposition was used,
13 rather than a traditional environmental indicator, as the direct metric for this GIS analysis of
14 ecological response.

15 **5.3.8 Uncertainty and Variability**

16 The analyses for the terrestrial nutrient enrichment case study were based on measured
17 data and model predictions that each contain a number of areas of uncertainty. For example,
18 characterizing NO_x and SO_x deposition includes uncertainties in monitoring instrumentation and
19 measurement protocols, as well as limitations in the spatial extent of existing monitoring
20 networks. Also, there are no “true” measurements of dry deposition. Geographic limitations in
21 monitoring led to reliance on CMAQ model predictions. CMAQ has its own uncertainties in
22 model formulation and in the inputs which drive the model’s simulation chemistry and transport
23 processes.

24 There are also uncertainties associated with the spatial resolutions of the measured and
25 modeled data used in this case study, as well as spatial and temporal variability associated with
26 measurement and modeling. Uncertainties are associated with gridding the NADP measurements
27 to 12 km resolution and the representativeness of 12 km data for characterizing deposition in this
28 case study area, particularly for the small sites of CSS as noted below. Specific areas of
29 uncertainty associated with this case study of CSS include the following:

- 1 ▪ CSS declines have been observed in the absence of fire when elevated nitrogen levels are
2 present; declines have also been observed in the absence of elevated nitrogen, but due to
3 fire. Therefore, there is still a need for quantifiable and predictive results to indicate the
4 pressure of each variable, as well as the pressure of the combined variables (if synergism
5 is present).
- 6 ▪ Many studies allude to a degradation of CSS by assessing species richness and abundance,
7 but it is not clear that indicators of CSS ecosystem health have been adequately explored.
- 8 ▪ Ongoing CSS experiments are beginning to show changes in CSS in response to elevated
9 nitrogen over relatively long periods of time (Allen, personal communication, 2008). The
10 incremental process may be occurring more slowly than previous field research
11 experiments have lasted, making the reasons for the decline appear variable or
12 imperceptible over the duration of a typical study.
- 13 ▪ At this point, CSS is fragmented into many relatively small parcels. The CMAQ 2002 data
14 is being modeled at 4-km resolution. When these 4-km data become available, there may
15 be a better sense of the relationship between the current distribution of CSS and
16 atmospheric nitrogen loads and fire threat.
- 17 ▪ Very little research exists regarding the effects of ozone on CSS. Although there is some
18 support that ozone is negatively correlated with CSS, the role has yet to be quantified or
19 consistently studied (Westman, 1981).
- 20 ▪ There is uncertainty in the relationship between current CSS distribution and the changing
21 climate.

22 Areas of uncertainty for MCF include the following:

- 23 ▪ The long-term consequences of increased nitrogen on conifers are unclear.
- 24 ▪ The effects of ozone for both MCF and lichens confound the effects of nitrogen.
- 25 ▪ The intermingling of fire and nitrogen cycling require additional research.
- 26 ▪ Research suggests that critical load benchmarks can decrease over time if the nitrogen
27 benchmark is exceeded for long periods of time because of decreasing nitrogen
28 efficiencies within nitrogen-saturated ecosystems (Fenn et al., 2008).

- 1 ▪ There remains considerable uncertainty in the potential response of soil carbon to increases
2 in total reactive nitrogen additions.

3 Although there are uncertainties in the data, models and techniques used for this case
4 study, the most applicable measurements and state-of-the-science models were used with
5 consideration for data and models' relative strengths and limitations.

6 **5.4 CONCLUSIONS**

7 This chapter has examined the sensitivity and effects of nutrient enrichment on aquatic
8 and terrestrial ecosystems, and, although a diverse array of U.S. ecosystems exist, exposure
9 levels at which negative effects are observed appear to be generally comparable to levels
10 identified in other sensitive U.S. ecosystems (benchmarks range from 1.5 to 30.5 kg N/ha/yr).
11 Enrichment benchmarks are also comparable to those found in the Aquatic Acidification and
12 Terrestrial Acidification case studies (see Chapter 4). Further consideration of these comparable
13 benchmarks can inform the decision-making process for mitigating terrestrial and aquatic
14 acidification and enrichment.

15 **5.5 REFERENCES**

- 16 Aber J.D., C.L. Goodale, S.V. Ollinger, M.L. Smith, A.H. Magill, M.E. Martin, R.A. Hallett, and
17 J.L. Stoddard. 2003. Is nitrogen deposition altering the nitrogen status of northeastern
18 forests? *Bioscience* 53:375–389.
- 19 Acharya, G., and L.L. Bennett. 2001. “Valuing Open Space and Land-Use Patterns in Urban
20 Watersheds.” *Journal of Real Estate Finance and Economics* 22(2/3):221-237.
- 21 Allen, E.B. 2008. Personal communication from E.B. Allen, U.S. Department of Agriculture
22 Forest Service.
- 23 Allen, E.B., P.E. Padgett, A. Bytnerowicz, and R. Minnich. 1998. *Nitrogen Deposition Effects on*
24 *Coastal Sage Vegetation of Southern California*. General Tech. Rep. PSW-GTR-166.
25 U.S. Department of Agriculture, Forest Service, Albany, CA.

- 1 Allen, E.B., P.J. Temple, A. Bytnerowicz, M.J. Arbaugh, A.G. Sirulnik, and L.E. Rao. 2007.
2 Patterns of understory diversity in mixed coniferous forests of Southern California
3 impacted by air pollution. *TheScientificWorldJournal* 7(S1):247–263.
- 4 Anderson, E. 1989. “Economic Benefits of Habitat Restoration: Seagrass and the Virginia Hard-
5 Shell Blue Crab Fishery.” *North American Journal of Fisheries Management* 9:140-149.
- 6 Anderson, N.J., I. Renberg, and U. Segerstrom. 1995. Diatom production responses to the
7 development of early agriculture in a boreal forest lake-catchment (Kassjon, Northern
8 Sweden). *Journal of Ecology* 83:809–822.
- 9 Baron, J.S. 2006. Hindcasting nitrogen deposition to determine ecological critical load.
10 *Ecological Applications* 16:433–439.
- 11 Baron, J.S., D.S. Ojima, E.A. Holland, and W.J. Parton. 1994. Analysis of nitrogen saturation
12 potential in Rocky Mountain tundra and forest: Implications for aquatic systems.
13 *Biogeochemistry* 27:61–82.
- 14 Baron, J.S., H.M. Rueth, A.M. Wolfe, K.R. Nydick, E.J. Allstott, J.T. Minear, and B. Moraska.
15 2000. Ecosystem responses to nitrogen deposition in the Colorado Front Range.
16 *Ecosystems* 3:352–368.
- 17 Bergström, A., and M. Jansson. 2006. Atmospheric nitrogen deposition has caused nitrogen
18 enrichment and eutrophication of lakes in the northern hemisphere. *Global Change*
19 *Biology* 12:635–643.
- 20 Bergström, A., P. Blomqvist, and M. Jansson. 2005. Effects of atmospheric nitrogen deposition
21 on nutrient limitation and phytoplankton biomass in unproductive Swedish lakes.
22 *Limnology and Oceanography* 50:987–994.
- 23 Bobbink, R. 1998. Impacts of tropospheric ozone and airborne nitrogenous pollutants on natural
24 and semi-natural ecosystems: a commentary. *New Phytologist* 139:161–168.

- 1 Bobbink, R., M. Hornung, and J.G.M. Roelofs. 1998. The effects of air-borne nitrogen pollutants
2 on species diversity in natural and semi-natural European vegetation. *Journal of Ecology*
3 86:717–738.
- 4 Bowman, W.D., J.R. Gartner, K. Holland, and M. Wiedermann. 2006. Nitrogen critical loads for
5 alpine vegetation and terrestrial ecosystem response: are we there yet? *Ecological*
6 *Applications* 16:1183–1193.
- 7 Bowman, W.D., T.A. Theodose, J.C. Schardt, and R.T. Conant. 1993. Constraints of nutrient
8 availability on primary production in two alpine tundra communities. *Ecology* 74:2085–
9 2097.
- 10 Bowman, W.D., J.R. Gartner, K. Holland, and M. Wiedermann. 2006. Nitrogen critical loads for
11 alpine vegetation and terrestrial ecosystem response: are we there yet? *Ecological*
12 *Applications* 16:1183–1193.
- 13 Boyer, E.W., C.L. Goodale, N.A. Jaworski, and R.W. Howarth. 2002. Anthropogenic nitrogen
14 sources and relationships to riverine nitrogen export in the northeastern U.S.A.
15 *Biogeochemistry* 57/58:137–169.
- 16 Bricker, S. 2008. Personal communication from Suzanne Bricker, National Oceanic and
17 Atmospheric Administration, to Jennifer Schimek, RTI.
- 18 Bricker, S., B. Buddemeier, S. Smith, B. Maxwell. In prep. *Results of Type Classification*
19 *Analyses from NOAA Workshop September, 2003 and KGS Work Session January, 2004.*
20 White paper. U.S. Department of Commerce, National Oceanic and Atmospheric
21 Administration, National Ocean Service, Silver Spring, MD.
- 22 Bricker, S., B. Longstaff, W. Dennison, A. Jones, K. Boicourt, C. Wicks, and J. Woerner. 2007.
23 *Effects of Nutrient Enrichment in the Nation's Estuaries: A Decade of Change.* NOAA
24 Coastal Ocean Program Decision Analysis Series No. 26. U.S. Department of Commerce,
25 National Oceanic and Atmospheric Administration, National Ocean Service, National
26 Centers for Coastal Ocean Science, Silver Spring, MD.

- 1 Bricker, S., D. Lipton, A. Mason, M. Dionne, D. Keeley, C. Krahforst, J. Latimer, and J.
2 Pennock. 2006. *Improving Methods and Indicators for Evaluating Coastal Water*
3 *Eutrophication: A Pilot Study in the Gulf of Maine*. NOAA Technical Memorandum
4 NOS NCCOS 20. U.S. Department of Commerce, National Oceanic and Atmospheric
5 Administration, National Ocean Service, National Centers for Coastal Ocean Science,
6 Center for Coastal Monitoring and Assessment, Silver Spring, MD.
- 7 Bricker, S.B., C.G. Clement, D.E. Pirhalla, S.P. Orlando, and D.R.G. Farrow. 1999. *National*
8 *Estuarine Eutrophication Assessment: Effects of Nutrient Enrichment in the Nation's*
9 *Estuaries*. NOAA—NOS Special Projects Office. U.S. Department of Commerce,
10 National Oceanic and Atmospheric Administration, National Ocean Service, National
11 Centers for Coastal Ocean Science, Center for Coastal Monitoring and Assessment,
12 Silver Spring, MD.
- 13 Bricker, S.B., J.G. Ferreira, and T. Simas. 2003. An integrated methodology for assessment of
14 estuarine trophic status. *Ecological Modelling* 169:39–60.
- 15 Brooks, P.D., M.W. Williams, and S.K. Schmidt. 1996. Microbial activity under alpine
16 snowpacks, Niwot Ridge, CO. *Biogeochemistry* 32:93–113.
- 17 Burger, J.C., R.A. Redak, E.B. Allen, J.T. Rotenberry, and M.F. Allen. 2003. “Restoring
18 Arthropod Communities in Coastal Sage Scrub.” *Conservation Biology* 17(2):460-467.
- 19 Burkholder, J.M., D.A. Dickey, C.A. Kinder, R.E. Reed, M.A. Mallin, M.R. McIver, L.B.
20 Cahoon, G. Melia, C. Brownie, J. Smith, N. Deamer, J. Springer, H.B. Glasgow, and D.
21 Toms. 2006. Comprehensive trend analysis of nutrients and related variables in a large
22 eutrophic estuary: A decadal study of anthropogenic and climatic influences. *Limnology*
23 *and Oceanography* 51:463–487.
- 24 Burns, D.A. 2004. The effects of atmospheric nitrogen deposition in the Rocky Mountains of
25 Colorado and southern Wyoming, USA--a critical review. *Environmental Pollution* 127:
26 57–269.

- 1 Bytnerowicz, A., M. Arbaugh, S. Schilling, W. Fraczek, D. Alexander, and P. Dawson. 2007. Air
2 pollution distribution patterns in the San Bernardino Mountains of Southern California: A
3 40-year perspective. DOI 10.1100/tsw.2007.57. *The Scientific World Journal* 7(S1):98–
4 109.
- 5 Bytnerowicz, A., and M.E. Fenn. 1996. Nitrogen deposition in California forests: a review
6 *Environmental Pollution* 92(2):127–146.
- 7 CAL FIRE (California Department of Forestry and Fire Protection). 1996. California Fire Plan.
8 Available at http://cdfdata.fire.ca.gov/fire_er/fpp_planning_cafireplan.
- 9 CAL FIRE (California Department of Forestry and Fire Protection). 2008. CAL FIRE 2007
10 Wildland Fire Summary.
- 11 California State Parks. 2003. Public Opinions and Attitudes on Outdoor Recreation in California
12 2002. California State Parks, Sacramento, California. December 2003.
- 13 Chesapeake Bay Executive Council. 2000.
14 http://www.chesapeakebay.net/content/publications/cbp_12081.pdf
- 15 CBP (Chesapeake Bay Program). 2009. *Chesapeake Bay Program: A Watershed Partnership*.
16 Online information. Chesapeake Bay Program, Annapolis, MD. Available at:
17 <http://www.chesapeakebay.net> (accessed February 2009).
- 18 Clark, C.M., and D. Tilman. 2008. Loss of plant species after chronic low-level nitrogen
19 deposition to prairie grasslands. *Nature* 451:712–715.
- 20 Das, B., R.D. Vinebrook, A. Sanchez-Azofeifa, B. Rivard, A.P. Wolfe. 2005. Inferring
21 sedimentary chlorophyll concentrations with reflectance spectroscopy: a novel approach
22 to reconstructing historical changes in the trophic status of mountain lakes. *Canadian*
23 *Journal of Fisheries and Aquatic Sciences* 62:1067-1078.
- 24 D’Elia, C.J., J.G. Sanders, and W.R. Boynton. 1986. Nutrient enrichment studies in a coastal
25 plain estuary: phytoplankton growth in large-scale, continuous cultures. *Canadian*
26 *Journal of Fisheries and Aquatic Sciences* 43:397–406.

- 1 Denning, A.S., J. Baron, M.A. Mast and M. Arthur. 1991. Hydrologic pathways and chemical
2 composition of runoff during snowmelt in Loch Vale Watershed, Rocky Mountain
3 National Park, Colorado, USA. *Water, Air, & Soil Pollution* 59:107-123.
- 4 Dortch, Q., and T.E. Whitledge. 1992. Does nitrogen or silicon limit phytoplankton production in
5 the Mississippi River plume and nearby regions? *Continental Shelf Research* 12:1293–
6 1309.
- 7 Eadie, B.J., B.A. McKee, M.B. Lansing, J.A. Robbins, S. Metz, and J.H. Trefry. 1994. Records
8 of nutrient-enhanced coastal productivity in sediments from the Louisiana continental
9 shelf. *Estuaries* 17:754–765.
- 10 Egerton-Warburton, L.M., and E.B. Allen. 2000. Shifts in arbuscular mycorrhizal communities
11 along an anthropogenic nitrogen deposition gradient. *Ecological Applications* 10(2):484–
12 496.
- 13 Egerton-Warburton, L.M., R.G. Graham, E.B. Allen, and M.F. Allen. 2001. Reconstruction of
14 the historical changes in mycorrhizal fungal communities under anthropogenic nitrogen
15 deposition. *Proceedings of the Royal Society of London B* 268:2479–2484.
- 16 Elser, J.J., M.E.S. Bracken, E.E. Cleland, D.S. Gruner, W.S. Harpole, H. Hillebrand II, J.T. Ngai,
17 E.W. Seabloom, J.B. Shurin, and J.E. Smith. 2007. Global analysis of nitrogen and
18 phosphorus limitation of primary producers in freshwater, marine, and terrestrial
19 ecosystems. *Ecological Letters* 10:1135–1142.
- 20 Elser, J.J., E.R. Marzolf, and C.R. Goldman. 1990. Phosphorus and nitrogen limitation of
21 phytoplankton growth in the freshwaters of North America: A review and critique of
22 experimental enrichments. *Canadian Journal of Fisheries and Aquatic Sciences* 47:1468–
23 1477.
- 24 Elser, JJ;Bracken, MES;Cleland, EE;Gruner, DS;Harpole, WS;Hillebrand, H;Ngai,
25 JT;Seabloom, EW;Shurin, JB;Smith, JE. 2007. Global analysis of nitrogen and
26 phosphorus limitation of primary producers in freshwater, marine and terrestrial
27 ecosystems. *ECOL LETT*, 10 (12): 1135-1142 DEC 2007

- 1 Enders, S.K., M. Pagani, S. Pantoja, J.S. Baron, A.P. Wolfe, N. Pedentchouk, L. Nuñez. 2008.
2 Compound-specific stable isotopes of organic compounds from lake sediments track
3 recent environmental changes in an alpine ecosystem, Rocky Mountain National Park,
4 United States of America) *Limnology and Oceanography* 53(4):1468-1478.
- 5 Fenn, M.E., L. Geiser, R. Bachman, T.J. Blubaugh, and A. Bytnerowicz. 2007. Atmospheric
6 deposition inputs and effects on lichen chemistry and indicator species in the Columbia
7 River Gorge, USA. *Environmental Pollution* 146:77–91.
- 8 Fenn, M.E., and A. Bytnerowicz. 1997. Summer throughfall and winter deposition in the San
9 Bernardino Mountains in southern California. *Atmospheric Environment* 31(5):673–683.
- 10 Fenn, M.E., J.W. Baron, E.B. Allen, H.M. Rueth, K.R. Nydick, L. Geiser, W.D. Bowen, J.O.
11 Sickman, T. Meixner, D.W. Johnson, and P. Neitlich. 2003. Ecological effects of
12 nitrogen deposition in the western United States. *Bioscience* 53(4):404–420.
- 13 Fenn, M.E., M.A. Poth, S.L. Schilling, and D.B. Grainger. 2000. Throughfall and fog deposition
14 of nitrogen and sulfur at an N-limited and N-saturated site in the San Bernardino
15 Mountains, Southern California. *Canadian Journal of Forest Research* 30:1476–1488.
- 16 Fenn, M.E., S. Jovan, F. Yuan, L. Geiser, T. Meixner, and B.S. Gimeno. 2008. Empirical and
17 simulated critical loads for nitrogen deposition in California mixed conifer forests.
18 *Environmental Pollution* 155(3):492–511.
- 19 Ferreira, J.G., S.B. Bricker, and T.S. Castro. 2007. Application and sensitivity testing of a
20 eutrophication assessment method on coastal systems in the United States and European
21 Union. *Journal of Environmental Management* 82:433–445.
- 22 Geiser, L.H., and P.N. Neitlich. 2007. Air pollution and climate gradients in Western Oregon and
23 Washington indicated by epiphytic macrolichens. *Environmental Pollution* 145:203–218.
- 24 Geoghegan, J., L.A. Wainger, and N.E. Bockstael. 1997. “Spatial Landscape Indices in a
25 Hedonic Framework: An Ecological Economics Analysis Using GIS.” *Ecological*
26 *Economics* 23:251-264.

- 1 Grulke N.E., C. Andersen, and W.E. Hogsett. 2001. Seasonal changes in above- and
2 belowground carbohydrate concentrations of ponderosa pine along a pollution gradient.
3 *Tree Physiology* 21:175–184.
- 4 Grulke, N.E., and L. Balduman. 1999. Deciduous conifers: high N deposition and O₃ exposure
5 effects on growth and biomass allocation in ponderosa pine. *Water, Air, and Soil*
6 *Pollution* 116:235–248.
- 7 Grulke, N.E., C.P. Andersen, M.E. Fenn, and P.R. Miller. 1998. Ozone exposure and nitrogen
8 deposition lowers root biomass of ponderosa pine in the San Bernardino Mountains,
9 California. *Environmental Pollution* 103:63–73.
- 10 Grulke, N.E., R.A. Minnich, T.D. Paine, S.J. Seybold, D. Chavez, M.E Fenn, P.J. Riggan, and A.
11 Dunn. 2008. Air pollution increases forest susceptibility to wildfires: a case study for the
12 San Bernardino Mountains in southern California. In *Wild Land Fires and Air Pollution*,
13 8. Edited by A. Bytnerowicz, M. Arbaugh, A. Riebau, and C. Andersen. Burlington, MA:
14 Elsevier.
- 15 Howarth, R.W., and R. Marino. 2006. Nitrogen as the limiting nutrient for eutrophication in
16 coastal marine ecosystems: evolving views over three decades. *Limnology and*
17 *Oceanography* 51:364–376.
- 18 Howarth, R. W.; Sharpley, A.; Walker, D. 2002a. Sources of nutrient pollution to coastal waters
19 in the United States: Implications for achieving coastal water quality goals. *Estuaries* 25:
20 656-676.
- 21
22 Howarth, R.W., E.W. Boyer, W.J. Pabich, and J.N. Galloway. 2002b. Nitrogen use in the United
23 States from 1961-2000 and potential future trends. *AMBIO: A Journal of the Human*
24 *Environment* 31:88–96.
- 25 Interlandi, S.J., and S.S. Kilham. 1998. Assessing the effects of nitrogen deposition on mountain
26 waters: a study of phytoplankton community dynamics. *Water Science and Technology*
27 38:139–146.

- 1 Irwin, E.G. 2002. “The Effects of Open Space on Residential Property Values.” *Land Economics*
2 78(4):465-480.
- 3 Jovan, S. 2008. *Lichen bioindication of biodiversity, air quality, and climate: baseline results*
4 *from monitoring in Washington, Oregon, and California*. General Technical Report.
5 PNW-GTR-737. U.S. Department of Agriculture, Forest Service, Pacific Northwest
6 Research Station, Portland, OR.
- 7 Jovan, S., and B. McCune. 2005. Air-quality bioindication in the greater central valley of
8 California, with epiphytic macrolichen communities. *Ecological Applications* 15:1712–
9 1726.
- 10 Justic, D., N.N. Rabalais, R.E. Turner, and Q. Dortch. 1995a. Changes in nutrient structure of
11 river-dominated coastal waters: Stoichiometric nutrient balance and its consequences.
12 *Estuarine, Coastal and Shelf Science* 40:339–356.
- 13 Justic, D., N.N. Rabalais, and R.E. Turner. 1995b. Stoichiometric nutrient balance and origin of
14 coastal eutrophication. *Marine Pollution Bulletin* 30:41–46.
- 15 Kahn, J.R., and W.M. Kemp. 1985. “Economic Losses Associated with the Degradation of an
16 Ecosystem: The Case of Submerged Aquatic Vegetation in Chesapeake Bay.” *Journal of*
17 *Environmental Economics and Management* 12:246-263.
- 18 Kaval, P., and J. Loomis. 2003. *Updated Outdoor Recreation Use Values With Emphasis On National*
19 *Park Recreation*. Final Report October 2003, under Cooperative Agreement CA 1200-99-009,
20 Project number IMDE-02-0070.
- 21 Keeler-Wolf, T. 1995. Post-fire emergency seeding and conservation in Southern California
22 shrublands. Pp. 127–139 in *Brushfires in California Wildlands: Ecology and Resource*
23 *Management*. Edited by J.E. Keeley and T. Scott. International Association of Wildland
24 Fire, Fairfield, WA.
- 25 Keeley, J.E., M.B. Keeley, and C.J. Fotheringham. 2005. Alien plant patterns during postfire
26 succession in Mediterranean-climate California shrublands. *Ecological Applications*
27 15(6):2109–2125.

- 1 Knowler, D. 2002. “A Review of Selected Bioeconomic Models: With Environmental Influences
2 in Fisheries.” *Journal of Bioeconomics* 4:163-181.
- 3 Lafrancois, B.M., K.R. Nydick, B.M. Johnson, and J.S. Baron. 2004. Cumulative effects of
4 nutrients and pH on the plankton of two mountain lakes. *Canadian Journal of Fisheries
5 and Aquatic Sciences* 61:1153–1165.
- 6 Land Trust Alliance. 2006. *The 2005 National Land Trust Census Report*. Washington, D.C.:
7 Land Trust Alliance, November 30, 2006.
- 8 Landers, D.H., J.M. Eilers, D.F. Brakke, W.S. Overton, P.E. Kellar, M.E. Silverstein, R.D.
9 Schonbrod, R.E. Crowe, R.A. Linthurst, J.M. Omernik, S.A. Teague, and E.P. Meier.
10 1987. *Western Lake Survey Phase I: Characteristics of Lakes in the Western United
11 States, Volume I: Population Descriptions and Physicochemical Relationships*.
12 EPA/600/3-86-054A. U.S. Environmental Protection Agency, Office of Acid Deposition,
13 Environmental Monitoring and Quality Assurance, Washington, DC.
- 14 Lipton, D. W. 1999. “Pfiesteria’s Economic Impact on Seafood Industry Sales and Recreational
15 Fishing.” In *Proceedings of the Conference, Economics of Policy Options for Nutrient
16 Management and Pfiesteria*. B. L. Gardner and L. Koch, eds., pp. 35–38. College Park,
17 MD: Center for Agricultural and Natural Resource Policy, University of Maryland.
- 18 Loomis, J., D. Griffin, E. Wu, and A. González-Cabán. 2002. “Estimating the Economic Value
19 of Big Game Habitat Production from Prescribed Fire Using a Time Series Approach.”
20 *Journal of Forest Economics* 2:119-29.
- 21 Loomis, J.B., and D.S. White. 1996. “Economic Benefits of Rare and Endangered Species:
22 Summary and Meta-Analysis.” *Ecological Economics* 18(3):197-206.
- 23 Lovett, G.M., and T.H. Tear. 2007. *Effects of Atmospheric Deposition on Biological Diversity in
24 the Eastern United States*. Workshop Report. Institute of Ecosystem Studies, Millbrook,
25 NY, and The Nature Conservancy, Albany, NY.

- 1 Mansfield, C.A., S.K. Pattanayak, W. McDow, R. MacDonald, and P. Halpin. 2005. “Shades of
2 Green: Measuring the Value of Urban Forests in the Housing Market.” *Journal of Forest
3 Economics* 11(3):177-199.
- 4 McKnight, D.M., R.L. Smith, J.P. Bradbury, J.S. Baron, and S. Spaulding. 1990. Phytoplankton
5 dynamics in three Rocky Mountain Lakes, Colorado U.S.A. *Arctic, Antarctic, and Alpine
6 Research* 22:264–274.
- 7 MEA (Millennium Ecosystem Assessment). 2005. *Ecosystems and Human Well-Being: Current
8 State and Trends: Findings of the Condition and Trends Working Group*. Edited by R.
9 Hassan, R. Scholes, and N. Ash. Washington, DC: Island Press.
- 10 Meixner, T., and M. Fenn. 2004. Biogeochemical budgets in a Mediterranean catchment with
11 high rates of atmospheric N deposition – importance of scale and temporal asynchrony.
12 *Biogeochemistry* 70:331–356.
- 13 Minnich, R.A., M.G. Barbour, J.H. Burk, and R.F. Fernav. 1995. Sixty years of change in conifer
14 forests of the San Bernardino Mountains: Reconstruction of California mixed conifer
15 forests prior to fire suppression. *Conservation Biology* 9:902–914. NC DENR (North
16 Carolina Department of Environment and Natural Resources). 2002. *Neuse River Basin-
17 wide Water Quality Plan*. North Carolina Department of Environment and Natural
18 Resources, Division of Water Quality/Planning, Raleigh, NC. Available at
19 <http://h2o.enr.state.nc.us/basinwide/Neuse/2002/Section A Chapter 2.pdf>.
- 20 Mistiaen, J.A., I.E. Strand, and D. Lipton. 2003. “Effects of Environmental Stress on Blue Crab
21 (*Callinectes sapidus*) Harvests in Chesapeake Bay Tributaries.” *Estuaries* 26(2a): 316-
22 322.
- 23 National Association of State Foresters (NASF). 2009. Quadrennial Fire Review
24 2009. Washington, DC: NASF. *Quadrennial Fire and Fuel Review Final Report 2009*.
25 National Wildfire Coordinating Group Executive Board January 2009.
- 26 National Oceanic and Atmospheric Administration (NOAA). 2007. “Annual Commercial
27 Landing Statistics.” Available at [http://www.st.nmfs.noaa.gov/
28 st1/commercial/landings/annual_landings.html](http://www.st.nmfs.noaa.gov/st1/commercial/landings/annual_landings.html).

- 1 North Carolina Department of Environment and Natural Resources (NC DENR). 2002.
2 Basinwide Planning Program : 2002 Neuse River Basinwide Water Quality Plan.
3 <http://h2o.enr.state.nc.us/basinwide/Neuse/2002/plan.htm>
- 4 NRC (National Research Council). 2000. *Clean Coastal Waters: Understanding and Reducing*
5 *the Effects of Nutrient Pollution*. Washington, DC: The National Academies Press.
- 6 Officer, C.B., R.B. Biggs, J.L. Taft, L.E. Cronin, M.A. Tyler, and W.R. Boynton. 1984.
7 Chesapeake Bay anoxia: Origin, development and significance. *Science* 223:22–27.
- 8 Padgett, P.E., and E.B. Allen. 1999. Differential responses to nitrogen fertilization in native
9 shrubs and exotic annuals common to Mediterranean coastal sage scrub of California.
10 *Plant Ecology* 144:93–101.
- 11 Padgett, P.E., E.B. Allen, A. Bytnerowicz, and R.A. Minnich. 1999. Changes in soil inorganic
12 nitrogen as related to atmospheric nitrogenous pollutants in southern California.
13 *Atmospheric Environment* 33:769–781.
- 14 Paerl, H., J. Pinckney, J. Fear, and B. Peierls. 1998. Ecosystem responses to internal and
15 watershed organic matter loading: Consequences for hypoxia in the eutrophying Neuse
16 River Estuary, NC, USA. *Marine Ecology Progress Series* 166:17–25.
- 17 Paerl, H.W., W.R. Boynton, R.L. Dennis, C.T. Driscoll, H.S. Greening, J.N. Kremer, N.N.
18 Rabalais, and S.P. Seitzinger. 2001a. Atmospheric deposition of nitrogen in coastal
19 waters: Biogeochemical and ecological implications. Pp. 11–52 in *Nitrogen Loading in*
20 *Coastal Water Bodies: An Atmospheric Perspective*. Coastal and Estuarine Series,
21 Volume 57. Edited by R.W. Valigura, R.B. Alexander, M.S. Castro, T.P. Meyers, H.W.
22 Paerl, P.E. Stacey, and R.E. Turner. Washington, DC: American Geophysical Union.
- 23 Paerl, H.W. 2002. Connecting atmospheric nitrogen deposition to coastal eutrophication.
24 *Environmental Science & Technology* 36:323A–326A.
- 25 Parsons, G., A.O. Morgan, J.C. Whitehead, and T.C. Haab. 2006. “The Welfare Effects of
26 Pfiesteria-Related Fish Kills: A Contingent Behavior Analysis of Seafood Consumers.”
27 *Agricultural and Resource Economics Review* 35(2):1-9.

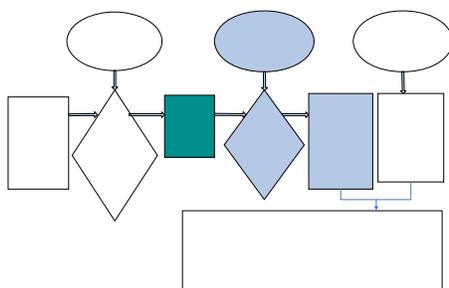
- 1 Preston, S.D., and J.W. Brakebill. 1999. *Applications of Spatially Referenced Regression*
2 *Modeling for the Evaluation of Total Nitrogen Loading in the Chesapeake Bay*
3 *Watershed*. USGS Water-Resources Investigations Report 99-4054. U.S. Department of
4 the Interior, U.S. Geological Survey, MD-DE-DC Water Science Center, Baltimore, MD.
5 Available at <http://md.usgs.gov/publications/wrir-99-4054>.
- 6 Reavie, E.D., and J.P. Smol. 2001. Diatom-environmental relationships in 64 alkaline
7 southwestern Ontario (Canada) lakes: a diatom-based model for water quality
8 reconstructions. *Journal of Paleolimnology* 25:27–44.
- 9 Renberg, I., T. Korsman, and N.J. Anderson. 1993. A temporal perspective of lake acidification
10 in Sweden. *AMBIO: a Journal of the Human Environment* 22:264–271.
- 11 Riegman, R. 1992. Phaeocystis blooms and eutrophication of the continental coastal zones of the
12 North Sea. *Marine Biology* 112:479–484.
- 13 Rueth, H.M., J.S. Baron, and E.J. Allstott. 2003. Responses of Engelmann spruce forests to
14 nitrogen fertilization in the Colorado Rocky Mountains. *Ecological Applications* 13:664–
15 673.
- 16 Saros, J.E., S.J. Interlandi, A.P. Wolfe, and D.R. Engstrom. 2003. Recent changes in the diatom
17 community structure of lakes in the Beartooth Mountain Range, USA. *Arctic, Antarctic,*
18 *and Alpine Research* 35:18–23.
- 19 Saros, J.E., T.J. Michel, S.J. Interlandi, and A.P. Wolfe. 2005. Resource requirements of
20 *Asterionella formosa* and *Fragilaria crotonensis* in oligotrophic alpine lakes:
21 implications for recent phytoplankton community reorganizations. *Canadian Journal of*
22 *Fisheries and Aquatic Sciences* 62:1681–1689.
- 23 Sherrod, S.K., and T.R. Seastedt. 2001. Effects of the northern pocket gopher (*Thomomys*
24 *talpoides*) on alpine soil characteristics, Niwot Ridge, CO. *Biogeochemistry* 55:195–218.
- 25 Sigal, L.L., and T.H. Nash, III. 1983. Lichen communities on conifers in southern California: an
26 ecological survey relative to oxidant air pollution. *Ecology* 64:1343–1354.

- 1 Siguenza, C., D.E. Crowley, and E.B. Allen. 2006. Soil microorganisms of a native shrub and
2 exotic grasses along a nitrogen deposition gradient in southern California. *Applied Soil*
3 *Ecology* 32:13–26.
- 4 Sirulnik, A.G., E.B. Allen, T. Meixner, and M.F. Allen. 2007. Impacts of anthropogenic N
5 additions on nitrogen mineralization from plant litter in exotic annual grasslands. *Soil*
6 *Biology and Biochemistry* 39:24–32.
- 7 Smith, R.A., and R.B. Alexander. 2000. Sources of nutrients in the nation’s watersheds. In
8 *Managing Nutrients and Pathogens from Animal Agriculture*. Proceedings from the
9 Natural Resource, Agriculture, and Engineering Service Conference for Nutrient
10 Management Consultants, Extension Educators, and Producer Advisors, Camp Hill, PA,
11 March 28–30. Natural Resource, Agriculture, and Engineering Service, Cooperative
12 Extension, Ithaca, NY.
- 13 Smith, V.K., C. Poulos, and H. Kim. 2002. “Treating Open Space as an Urban Amenity.”
14 *Resource and Energy Economics* 24:107-129.
- 15 Spruill, T.B., A.J. Tesoriero, H.E. Mew, Jr., K.M. Farrell, S.L. Harden, A.B. Colosimo, and S.R.
16 Kraemer. 2004. Scientific Investigations Report 2004-5283. U.S. Department of the
17 Interior, U.S. Geological Survey, Reston, VA. Available at
18 <http://pubs.usgs.gov/sir/2004/5283/>.
- 19 Stanley, D.L. 2005. Local Perceptions of Public Goods: Recent Assessments of Willingness-to-
20 Pay for Endangered Species. *Contemporary Economic Policy*, Vol 23, No. 2, April 2005,
21 165-179.
- 22 Steltzer, H., and W.D. Bowman. 1998. Differential influence of plant species on soil nitrogen
23 transformations within moist meadow alpine tundra. *Ecosystems* 1:464–474.
- 24 Sterner, R.W., and J.J. Elser. 2002 *Ecological Stoichiometry: The Biology of Elements from*
25 *Molecules to the Biosphere*. Princeton, NJ: Princeton University Press.
- 26 Stoddard, J., J.S. Kahl, F.A. Deviney, D.R. DeWalle, C.T. Driscoll, A.T. Herlihy, J.H. Kellogg,
27 P.S. Murdoch, J.R. Webb, and K.E. Webster. 2003. *Response of Surface Water Chemistry*

- 1 to the Clean Air Act Amendments of 1990. EPA 620/R-03/001. U.S. Environmental
2 Protection Agency, Office of Research and Development, National Health and
3 Environmental Effects Research Laboratory, Research Triangle Park, NC.
- 4 Suding, K.N., A.E. Miller, H. Bechtold, and W.D. Bowman. 2006. The consequence of species
5 loss on ecosystem nitrogen cycling depends on community compensation. *Oecologia*
6 149:141–149.
- 7 Takemoto, B.K., A. Bytnerowicz, and M.E. Fenn. 2001. Current and future effects of ozone and
8 atmospheric nitrogen deposition on California’s mixed conifer forests. *Forest Ecology*
9 *and Management* 144:159–173.
- 10 Tank, J.L., and W.K. Dodds. 2003. Nutrient limitation of epilithic and epixylic biofilms in ten
11 North American streams. *Freshwater Biology* 48:1031–1049.
- 12 Turner, R.E., N.Qureshi, N.N. Rabalais, Q. Dortch, D. Justic, R.F. Shaw, and J. Cope. 1998.
13 Fluctuating silicate: Nitrate ratios and coastal plankton food webs. *Proceedings of the*
14 *National Academy of Sciences of the United States of America* 95:13048–13051.
- 15 Tyrvaainen, L., and A. Miettinen. 2000. Property prices and urban forest amenities. *Journal of*
16 *Environmental Economics and Management* 39(2):205–223.
- 17 U.S. DOI (Department of the Interior). 2007. *2006 National Survey of Fishing, Hunting, and*
18 *Wildlife-Associated Recreation*. U.S. Department of the Interior, Fish and Wildlife
19 Service, and U.S. Department of Commerce, U.S. Census Bureau. Washington, DC.
- 20 U.S. EPA (Environmental Protection Agency). 2000a. *Deposition of Air Pollutants to the Great*
21 *Waters. Third Report to Congress*. EPA-453/R-00-005. U.S. Environmental Protection
22 Agency, Office of Air Quality Planning and Standards, Research Triangle Park, NC.
23 Available at <http://www.epa.gov/air/oaqps/gr8water/3rdrp/> (accessed January 16, 2008).
- 24 U.S. EPA (Environmental Protection Agency). 2000b. *Guidelines for Preparing Economic*
25 *Analyses*. U.S. Environmental Protection Agency, Office of the Administrator,
26 Washington, DC. EPA 240-R-00-003.

- 1 U.S. EPA (Environmental Protection Agency). 2002. *Summary Table for Nutrient Criteria*
2 *Documents*. U.S. Environmental Protection Agency, Office of Water, Washington, DC.
3 Available at
4 <http://www.epa.gov/waterscience/criteria/nutrient/ecoregions/files/sumtable.pdf>.
- 5 U.S. EPA (Environmental Protection Agency). 2008. *Integrated Science Assessment (ISA) for*
6 *Oxides of Nitrogen and Sulfur–Ecological Criteria (Final Report)*. EPA/600/R-
7 08/082F. U.S. Environmental Protection Agency, National Center for Environmental
8 Assessment–RTP Division, Office of Research and Development, Research Triangle
9 Park, NC. Available at <http://cfpub.epa.gov/ncea/cfm/recordisplay.cfm?deid=201485>.
- 10 Vourlitis, G.L., G. Zorba, S.C. Pasquini, and R. Mustard. 2007. Carbon and nitrogen storage in
11 soil and litter of southern Californian semi-arid shrublands. *Journal of Arid Environments*
12 70:164–173.
- 13 Webster, E., and D. Mackay, 2003. *Defining Uncertainty and Variability in Environmental Fate*
14 *Models*. CEMC Report No. 200301. Canadian Environmental Modelling Centre, Trent
15 University, Peterborough, ON, Canada. Available at
16 www.trentu.ca/cemc/CEMC200301.pdf.
- 17 Weiss, S.B. 1999. Cars, cows, and checkerspot butterflies: Nitrogen deposition and management
18 of nutrient-poor grasslands for a threatened species. *Conservation Biology* 13:1476–1486.
- 19 Westman, W.E. 1981. Diversity relations and succession in Californian coastal sage scrub.
20 *Ecology* 62:170–184.
- 21 Whitall, D., and H.W. Paerl. 2001. Spatiotemporal Variability of wet atmospheric nitrogen
22 deposition to the Neuse River Estuary, North Carolina. *Journal of Environmental Quality*
23 30:1508–1515.
- 24 Whitall, D., S. Bricker, J. Ferreira, A.M. Nobre, T. Simas, and M. Silva. 2007. Assessment of
25 eutrophication in estuaries: Pressure-state-response and nitrogen source apportionment.
26 *Environmental Management* 40:678–690.

- 1 Whitehead, J.C., T.C. Haab, and G.R. Parsons. 2003. “Economic Effects of Pfiesteria.” *Ocean &*
2 *Coastal Management* 46(9-10):845-858.
- 3 Williams, M.W., J.S. Baron, N. Caine, R. Sommerfeld, and J.R. Sanford. 1996. Nitrogen
4 saturation in the Rocky Mountains. *Environmental Science and Technology* 30: 640–646.
- 5 Williams, M.W. and K.A. Tonnessen. 2000 Critical loads for inorganic nitrogen deposition in the
6 Colorado Front Range, USA. *Ecological Applications* 10:1648–1665.
- 7 Wolfe, A.P., J.S. Baron, and R.J. Cornett, 2001. Unprecedented changes in alpine ecosystems
8 related to anthropogenic nitrogen deposition. *Journal of Paleolimnology* 25:1-7.
- 9 Wolfe, A.P., Van Gorp, A.C. & J.S. Baron, 2003. Recent ecological and bioecological changes
10 in alpine lakes of Rocky Mountain National Park (Colorado, U.S.A.): a response to
11 anthropogenic nitrogen deposition. *Geobiology* 1: 153-168.
- 12 Wood, Y., T. Meixner, P.J. Shouse, and E.B. Allen. 2006. Altered Ecohydrologic response
13 drives native shrub loss under conditions of elevated N-deposition. *Journal of*
14 *Environmental Quality* 35:76–92.
- 15

1
2

3

6.0 ADDITIONAL EFFECTS

6.1 VISIBILITY, CLIMATE, AND MATERIALS

4 The Clean Air Act (CAA) definition of welfare effects includes, but is not limited to,
5 effects on soils, water, wildlife, vegetation, visibility, weather, and climate, as well as effects on
6 man-made materials, economic values, and personal comfort and well-being. This Risk and
7 Exposure Assessment focuses primarily on ecological effects resulting from current deposition
8 of compounds containing nitrogen and sulfur. Acidification (from both sulfur and nitrogen) and
9 nutrient enrichment (from nitrogen) are the central ecological effects addressed in this Risk and
10 Exposure Assessment (see Chapters 4 and 5). The additional welfare effects addressed in this
11 chapter include the influence of sulfur oxides (SO_x) deposition effects on mercury methylation,
12 nitrous oxide (N₂O) effects on climate, deposition effects of nitrogen oxides (NO_x) on biogenic
13 greenhouse gas fluxes, and phytotoxic effects on plants. While a quantitative assessment of these
14 important effects is beyond the scope of this review, this chapter will evaluate them qualitatively.
15

16 It is understood that impairment of visibility and materials damage can result from
17 atmospheric particulate matter (PM), which is composed in part of sulfate (SO₄²⁻)- and nitrate
18 (NO₃⁻)-based particulates (i.e., ammonium sulfate [(NH₄)₂SO₄] and ammonium nitrate
19 [NH₄NO₃]). As early as the 1982 *Air Quality Criteria for Particulate Matter and Sulfur Oxides*
20 (PM/SO_x Air Quality Criteria Document [AQCD]) document (U.S. EPA, 1982b), EPA has
21 documented that particulates contribute to the impairment of visibility. That document stated that
22 theoretical and empirical findings suggest that sulfates often dominate the fine particle mass and,
23 hence, impairment of visibility. It also acknowledged that particulate species of nitrates are
24 important as well, and it further stated that reductions in visibility can adversely affect
25 transportation safety, property values, and aesthetics. Materials damage, such as accelerated

1 corrosion of metal, erosion, soiling of paint, and soiling of buildings and other structures has
2 been documented. Since these effects are largely considered to be PM effects, they are being
3 addressed in the PM National Ambient Air Quality Standards (NAAQS) review currently
4 underway (See http://www.epa.gov/ttn/naaqs/standards/pm/s_pm_index.html for documents
5 related to that review).

6 **6.2 SULFUR AND MERCURY METHYLATION**

7 Behavioral, reproductive, neurochemical, and hormonal effects due to mercury have been
8 demonstrated in fish and in piscivorous mammals and birds (U.S. EPA, 1996; Scheuhammer
9 et al., 2007). Methylmercury has been shown to be the mercury compound that accumulates in
10 the tissues of affected fish and piscivorous species (Becker and Bigham, 1995; Bloom, 1992;
11 Harris et al., 2003; Scheuhammer et al., 2007). The production of the large majority of
12 methylmercury is mediated by sulfate-reducing bacteria (SRB), and changes in SO_4^{2-} deposition
13 have resulted in changes in both mercury methylation and mercury concentrations in fish.

15 **6.2.1 Science Background**

17 It is stated in the *Integrated Science*
19 *Assessment (ISA) for Oxides of Nitrogen and*
21 *Sulfur—Ecological Criteria (Final Report)* (ISA)

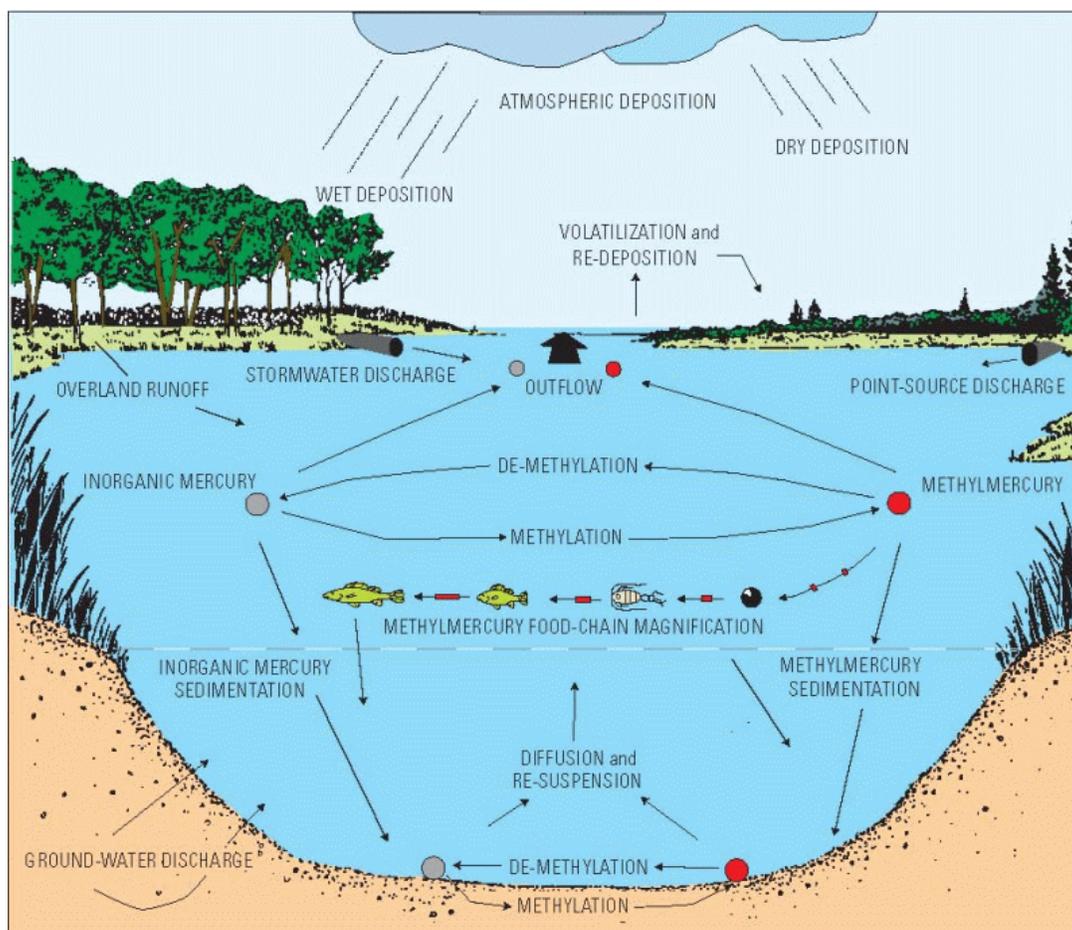
Current evidence indicates that in watersheds where mercury is present, increased SO_x deposition very likely results in methylmercury accumulation in fish (Munthe et al, 2007; Drevnick et al., 2007).

22 (U.S. EPA, 2008, Sections 3.4.1 and 4.5) that mercury is a highly neurotoxic contaminant that
23 enters the food web as a methylated compound, methylmercury. The contaminant is concentrated
24 in higher trophic levels, including fish eaten by humans. Experimental evidence has established
25 that only inconsequential amounts of methylmercury can be produced in the absence of SO_4^{2-} .
26 Many variables influence how much mercury accumulates in fish, but elevated mercury levels in
27 fish can only occur where substantial amounts of methylmercury are present. Current evidence
28 indicates that in watersheds where mercury is present, increased SO_x deposition very likely
29 results in methylmercury accumulation in fish (Drevnick et al., 2007; Munthe et al, 2007).

30 Establishing the quantitative relationship between SO_4^{2-} and mercury methylation in
31 natural settings is difficult because of the presence of multiple interacting factors in aquatic
32 environments where SO_4^{2-} , SRBs, and mercury are present. The amount of methylmercury
33 produced by bacteria varies with surface water oxygen content, temperature, pH, and supply of
34 labile organic carbon. When these interacting factors are outside of the ranges most favorable for

1 methylation, increasing levels of SO_4^{2-} deposition will not increase the amount of methylmercury
2 in the aquatic environment. For example, effects on mercury methylation in high-altitude lakes in
3 the Western United States have been recorded with changes in SO_4^{2-} deposition, where some of
4 those interacting factors were also outside of the ranges most favorable for methylation (U.S.
5 EPA, Sections 3.4 and 4.5). Watersheds with conditions known to be conducive to mercury
6 methylation have been identified in the northeastern United States and southeastern Canada
7 (Chen et al., 2005; Evers et al., 2007; Scheuhammer and Blancher, 1994; Scheuhammer et al.,
8 2007), whereas watersheds with elevated methylmercury levels observed in water or in fish are
9 seen in most of the continental United States.

10 Several interrelated factors seem to affect mercury uptake in fish, including low lake-
11 water pH, dissolved organic carbon, and suspended PM concentrations in the water column
12 (Driscoll et al., 1994; Grieb et al., 1990; Kamman et al., 2004; Mierle and Ingram, 1991; Sun
13 and Hitchin, 1990; U.S. EPA, 1996). For example, a lower pH can increase the ability of
14 methylmercury to permeate fish membranes and speed the rate of uptake, thus increasing
15 mercury residues in fish (Weiner et al., 2003). In addition, phosphorus and nitrogen can be
16 important as these factors regulate aquatic productivity and, thus, mercury concentrations in
17 aquatic organisms (Driscoll et al., 2001). The proportion of upland to wetland land area within a
18 watershed, as well as wetland type and annual water yield, also appears to be important (St.
19 Louis et al., 1996). **Figure 6.2-1** shows the process of mercury methylation in an aquatic
20 environment.

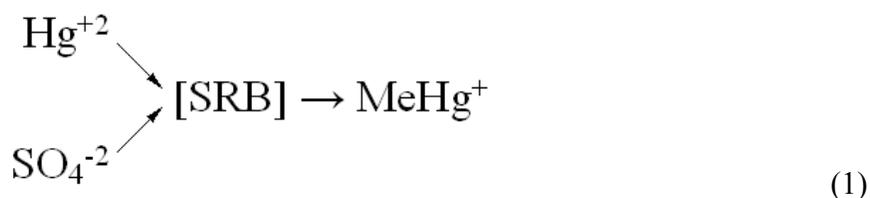


1
2 **Figure 6.2-1.** The mercury cycle in an ecosystem (USGS, 2006).

3 **6.2.2 Qualitative Analysis**

4 The role of atmospherically deposited sulfur species in mercury methylation varies
5 greatly across ecosystems. Field studies have determined that the majority of mercury
6 methylation occurs within anoxic waters and sediments (Gilmour et al., 1998; Hammerschmidt et
7 al., 2004; Watras et al., 1995); however, several studies have observed that quantitative
8 prediction of mercury methylation is impeded by the presence of multiple known interacting
9 factors whose influence on methylation has not been quantified. These include types of SRB,
10 sulfur species, mercury species, pH, organic acids, and other factors (Benoit et al., 2003;
11 Gilmour et al., 1992; Langer et al., 2001; Munthe et al., 2007; Watras and Morrison, 2008).
12 Methylation via iron-reducing bacteria has also been observed in anoxic, iron-rich sediments;
13 however, this process is not well understood and appears to be less extensive than the SRB-
14 mediated mercury methylation (Fleming et al., 2006; Kerin et al., 2006).

1 Methyl mercury output by SRBs is a by-product of the conversion of SO_4^{2-} to sulfide
 2 (Benoit et al., 2003; Branfireun et al., 1999; Compeau and Bartha, 1985; Gilmour et al., 1992). In
 3 general, the rate of methylmercury generation depends on the factors that affect SRB propagation
 4 and activity, the availability of inorganic mercury, and the demethylation of mercury. The
 5 introduction of SO_4^{2-} to SRB in the presence of divalent mercury (Hg^{+2}), usually in low oxygen
 6 sediments, leads to the following biomediated transformation:

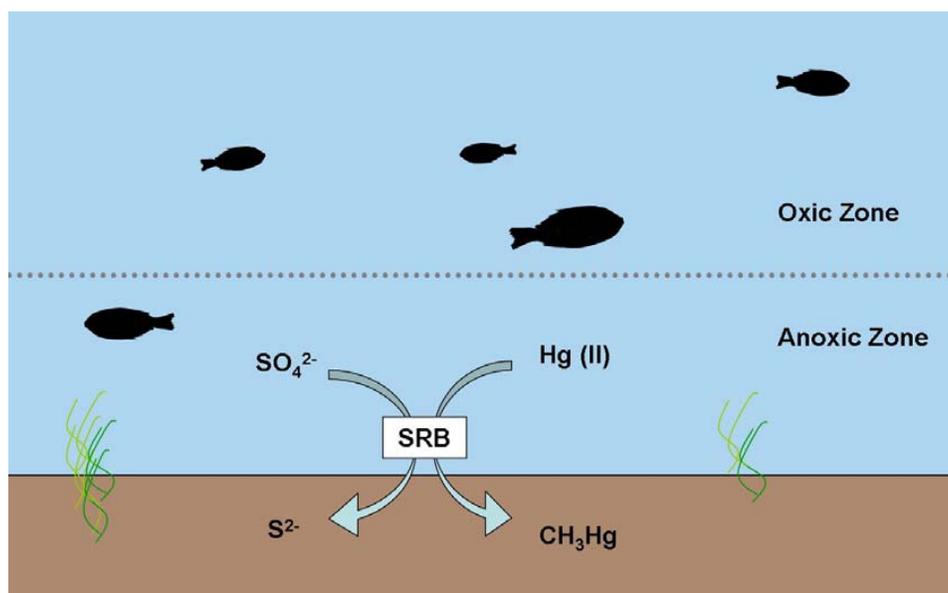


8 The presence of SO_4^{2-} , inorganic mercury, and SRB are, thus, the primary requirements
 9 for bacterially mediated sulfate-reducing mercury conversion. Additional factors affecting
 10 conversion include the presence of anoxic conditions, temperature, the presence and types of
 11 organic matter, the presence and types of mercury-binding species, and watershed effects (e.g.,
 12 watershed type, land cover, waterbody limnology, runoff loading). Demethylation, which
 13 involves aerobic and anaerobic microbial processes, as well as sunlight-dependent processes
 14 (e.g., photodemethylation), can also have a substantial effect; therefore, increased methylation in
 15 natural environments should be understood as increased *net* mercury methylation (Benoit et al.,
 16 2003).

17 The role of SO_4^{2-} in mercury methylation has been confirmed through a series of
 18 independent and interdependent studies. As noted in the ISA, early studies on Little Rock Lake,
 19 WI, first observed the link between sulfur enrichment, acidification, and methylmercury
 20 concentrations (Hrabik and Watras, 2002). Other important studies include Branfireun et al.
 21 (1999) and Jeremiason et al. (2006). The beneficial effect of decreased SO_4^{2-} deposition on fish
 22 tissue methylmercury concentrations has also recently been observed in an isolated Lake
 23 Superior ecosystem, where fish tissue concentrations fell below fish consumption advisory levels
 24 in the absence of any change in atmospheric mercury deposition (Drevnick et al., 2007). Other
 25 studies have focused on the biogeochemical process of mercury cycling to determine factors that
 26 are responsible for the link between methylmercury and acidification. Early research by Faust
 27 and Osman (1981) estimated that 90% to 99% of the total mercury concentration in surface
 28 waters was associated with sediment. With regard to methylmercury, the highest concentrations

1 in the environment generally occur at or near the sedimentary surface, below the oxic–anoxic
 2 boundary. The formation of methylmercury has also been associated with macrophytic
 3 vegetation and periphyton (Mauro et al., 2002). Mercury methylation rate and organic carbon
 4 substrates (e.g., acetate, lactate) may fluctuate when associated with the presence of SRB and
 5 environmental conditions (Mitchell et al., 2008). **Figure 6.2-2** illustrates the general SRB
 6 methylation process. It should be noted that mercury can also be supplied from sediments.

7 Although mercury methylation can occur within the water column, there is generally a far
 8 greater contribution of mercury methylation from sediments because of anoxia and of greater
 9 concentrations of SRB, substrate, and SO_4^{2-} . The conditions within sediment pores and
 10 conditions affecting sediment porewater may, therefore, play a key role in mercury methylation.
 11 The relative contribution of methylmercury from porewater in the surficial sediment layer is
 12 dependent on the size of the hypolimnetic anoxic zone, the location of the bacterioplankton
 13 activity, and several other factors, such as temperature, organic carbon content, and the presence
 14 of sulfides (Watras et al., 1995).



15
 16 **Figure 6.2-2.** Biogeochemical process of mercury methylation.

17 **6.2.2.1 Watershed Influences**

18 The effect of watersheds on methylmercury production is dependent on many factors
 19 (e.g., dissolved organic carbon, temperature, anoxia, SO_4^{2-}); however, watershed influences also
 20 include physical, chemical, and ecological variables that, in turn, have an impact on those

1 factors; they include land cover, precipitation response, hydrology, nutrient loading, and
2 limnology. Watershed influences may also play a role in the uptake of methylmercury into fish
4 and other aquatic species.

6 Land cover and land use affect the transport of
8 chemical species, such as mercury, nutrients, and
10 dissolved organic carbon. Methylmercury production
12 generally increases with increasing proportion of
13 wetlands in the contributing area to surface water systems (Benoit et al., 2003; Watras and
14 Morrison, 2008). In general, wetland environments tend to promote mercury methylation
15 because of increased anoxic environments, fresh organic matter, moderated temperature, and
16 macrophytic environments for bacterial activity (Back et al., 2002). Additionally, increased
17 forest cover and mixed agriculture have been correlated with increased mercury methylation in
18 downstream surface waters, presumably due to organic matter (Driscoll et al., 2007; Krabbenhoft
19 et al., 1999). Land disturbance may also contribute to increased mercury methylation
20 downstream by increasing erosion and, therefore, the mobility of mercury and organic matter
22 (Driscoll et al., 2007).

Methylmercury production generally increases with increasing proportion of wetlands in the area contributing to surface water systems (Benoit et al., 2003; Watras and Morrison, 2008).

24 State-level fish consumption advisories for
26 mercury are based on state criteria, many of which are
28 based on EPA's fish tissue criterion for methylmercury at
30 0.3 microgram per gram ($\mu\text{g/g}$) or on U.S. Food and Drug
31 Administration action limits of 1.0 mg/kg, equal to 1ppm by weight. Fish tissue concentrations
32 of methylmercury at this level or above are extremely unlikely to be observed without substantial
33 methylating activity in the watershed affected. There were 2,436 fish consumption advisories
34 across the United States in 2004; 2,682 in 2005; and 3,080 in 2006. Forty-eight states, one
35 territory, and two tribes have issued mercury advisories. Eighty percent of all fish consumption
36 advisories have been issued, at least in part, because of mercury. In 2006, a total of 14,177,175
37 lake acres and 882,963 river miles were under advisory for mercury (U.S. EPA, 2007a). **Figure**
38 **6.2-3** summarizes the spatial distribution patterns by state for documented fish consumption
39 advisory listings.

There were 3,080 fish consumption advisories in the United States in 2006, with 48 states, one territory, and two tribes having mercury advisories.

6.2.2.2 Conclusions

The ISA concluded that evidence is sufficient to infer a casual relationship between sulfur deposition and increased mercury methylation in wetlands and aquatic environments. There appears to be a relationship between SO_4^{2-} deposition and mercury methylation; however, the rate of mercury methylation varies according to several spatial and biogeochemical factors whose influence has not been fully quantified (see **Figure 6.2-4**). Therefore, the correlation between SO_4^{2-} deposition and methylmercury could not be quantified for the purpose of interpolating the association across waterbodies or regions. Nevertheless, because changes in methylmercury in ecosystems represent changes in significant human and ecological health risks, the association between sulfur and mercury cannot be neglected (U.S. EPA, 2008, Sections 3.4.1 and 4.5).

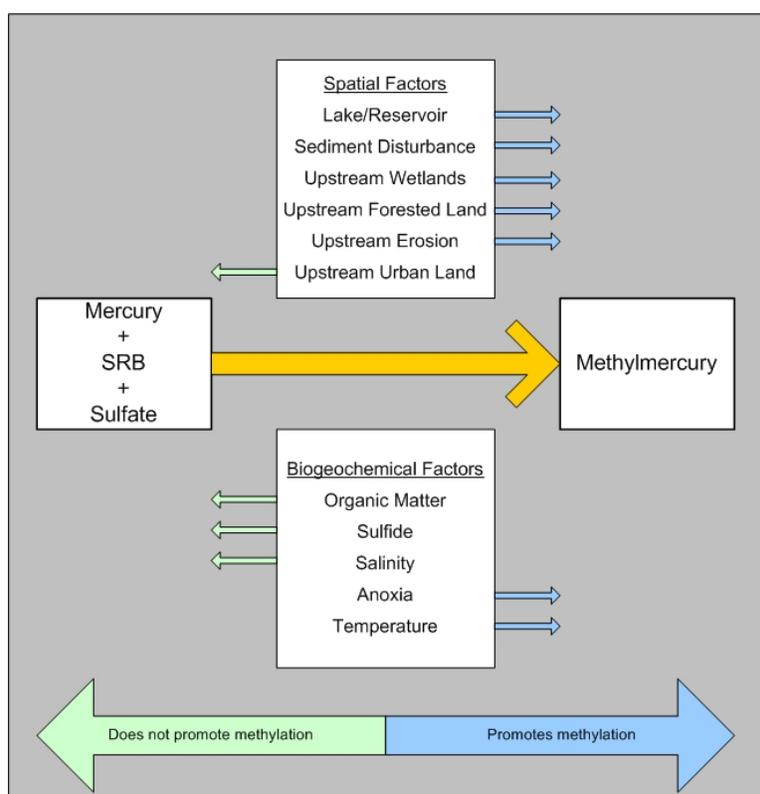


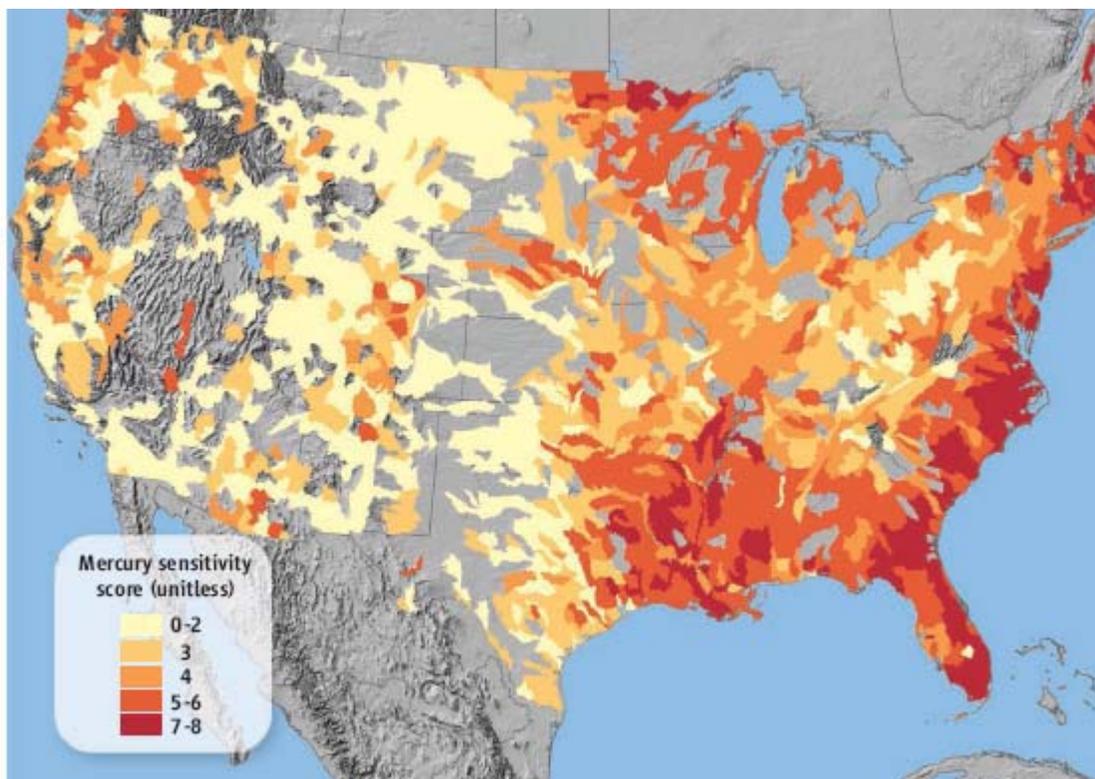
Figure 6.2-4. Spatial and biogeochemical factors influencing methylmercury production.

As research evolves and the computational capacity of models expands to meet the complexity of mercury methylation processes in ecosystems, the role of interacting factors may be better parsed out to identify ecosystems or regions that are more likely to generate higher

1 concentrations of methylmercury. **Figure 6.2-5** illustrates the type of current and forward-
2 looking research being developed by the U.S. Geological Survey (USGS) to synthesize the
3 contributing factors of mercury and to develop a map of sensitive watersheds. The mercury score
4 referenced in **Figure 6.2-5** is based on SO_4^{2-} concentrations, acid neutralizing capacity (ANC),
5 levels of dissolved organic carbon and pH, mercury species concentrations, and soil types to
6 gauge the methylation sensitivity (Myers et al., 2007).

8 Interdependent biogeochemical factors preclude the
10 existence of simple sulfate-related mercury methylation
12 models (see **Figure 6.2-4**). It is clear that decreasing sulfate
13 deposition is likely to result in decreased methylmercury concentrations. Future research may
14 allow for the characterization of a usable sulfate-methylmercury response curve; however, no
15 regional or classification calculation scale can be created at this time because of the number of
16 confounding factors.

It is evident that decreases in sulfate deposition will likely result in decreases in methylmercury concentration.



17
18 **Figure 6.2-5** Preliminary USGS map of mercury methylation-sensitive watersheds
19 derived from more than 55,000 water quality sites and 2,500 watersheds
20 (Myers et al., 2007).

1 Decreases in SO_4^{2-} emissions have already shown promising reductions in
2 methylmercury. Observed decreases in methylmercury fish tissue concentrations have been
3 linked to decreased acidification and declining SO_4^{2-} and mercury deposition in Little Rock
4 Lake, WI (Hrabik and Watras, 2002), and to decreased SO_4^{2-} deposition in Isle Royale in Lake
5 Superior, MI (Drevnick et al., 2007). Although the possibility exists that reductions in SO_4^{2-}
6 emissions could generate a pulse in methylmercury production because of decreased sulfide
7 inhibition in sulfate-saturated waters, this effect would likely involve a limited number of U.S.
8 waters (Harmon et al., 2007). Also, because of the diffusion and outward flow of both mercury-
9 sulfide complexes and SO_4^{2-} , increased mercury methylation downstream may still occur in
10 sulfate-enriched ecosystems with increased organic matter and/or downstream transport
11 capabilities.

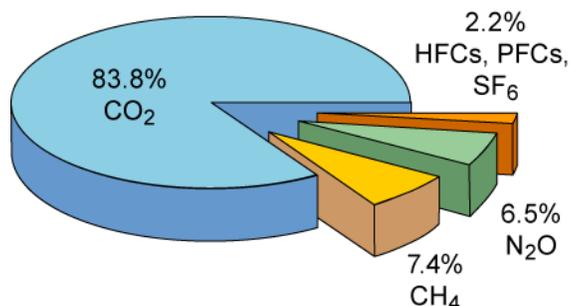
12 Remediation of sediments heavily contaminated with mercury has yielded significant
13 reductions of methylmercury in biotic tissues. Establishing quantitative relations in biotic
14 responses to methylmercury levels as a result of changes in atmospheric mercury deposition,
15 however, presents difficulties because direct associations can be confounded by all of the factors
16 discussed in this section. Current research does suggest that the levels of methylmercury and
17 total mercury in ecosystems are positively correlated, so that reductions in mercury deposited
18 into ecosystems would also eventually lead to reductions in methylmercury in biotic tissues.
19 Ultimately, an integrated approach that involves the reduction of both sulfur and mercury
20 emissions may be most efficient because of the variability in ecosystem responses. Reducing SO_x
21 could have a beneficial effect on levels of methylmercury in many waters of the United States.
22 This will be addressed, as appropriate, in the policy assessment portion of this review.

23 **6.3 NITROUS OXIDE**

24 Nitrous oxide (N_2O) has not been considered in setting previous nitrogen dioxide (NO_2)
25 NAAQS. In the first NO_x review, N_2O was not considered an air contaminant because there was
26 “no evidence to suggest N_2O is involved in photochemical reactions in the lower atmosphere”
27 (U.S. EPA, 1971). Nitrous oxide was addressed in both the 1982 and 1993 *Air Quality Criteria*
28 *for Oxides of Nitrogen* (NO_x AQCD) documents (U.S. EPA, 1982a, 1993). In 1982, it was
29 described as one of the eight nitrogen oxides that may be present in the ambient air, but “not
30 generally considered a pollutant.” The effect of N_2O on stratospheric ozone was described, and

1 the 1982 NO_x AQCD noted that N₂O may cause a small decrease in stratospheric ozone (U.S.
2 EPA, 1982a). Finally, the 1982 NO_x AQCD concluded that N₂O significantly contributes to the
3 atmospheric greenhouse effect by trapping outgoing terrestrial radiation, and that the issue was
4 being investigated, but that many years of research were still needed to assess the issue reliably
5 (U.S. EPA, 1982a). The 1993 NO_x AQCD also identified N₂O as an oxidized nitrogen compound
6 that is not generally considered to be an air pollutant, but it does have an impact on stratospheric
7 ozone and is considered to be among the more significant greenhouse gases (GHGs) (U.S. EPA,
8 1993). Although not considered within the scope of the previous review, these documents clearly
9 considered N₂O to be within the scope of the listed nitrogen oxides' criteria for pollutants.

10 The current ISA (U.S. EPA, 2008, Section 2.2) acknowledges that N₂O is a potent GHG
11 and discusses N₂O sources and emissions in the United States, as well as the biogeochemistry of
12 N₂O's microbial-mediated production via denitrification and nitrification in natural ecosystems.
13 Based on the current U.S. GHG inventory (U.S. EPA, 2007b), N₂O contributes approximately
14 6.5% to total GHG emissions (in carbon dioxide [CO₂] equivalents) (**Figure 6.3-1**).



15
16 **Figure 6.3-1.** Percentage of total U.S. emissions of greenhouse gases
17 in CO₂ equivalents (U.S. EPA, 2007b).

18 Since the definition of “welfare effects” includes effects on climate [CAA Section
19 302(h)], N₂O is included within the scope of this review. However, it is most appropriate to
20 analyze the role of N₂O in anthropogenic climate change in the context of all of the GHGs.
21 Because such an analysis is outside the scope of this review, it will not be a quantitative part of
22 this assessment.

23 Although the atmospheric concentration of N₂O (319 parts per billion in 2005) is much
24 lower than CO₂ (379 parts per million in 2005), its global warming potential is 296 times that of
25 CO₂. Human activities have increased the atmospheric concentration of N₂O by 18% since

1 preindustrial times (IPCC, 2007). The continuing increase of those GHG concentrations has been
2 shown to threaten human and ecosystem health (IPCC, 2007).

3 **6.4 NITROGEN ADDITION EFFECTS ON PRIMARY PRODUCTIVITY** 4 **AND BIOGENIC GREENHOUSE GAS FLUXES**

5 **6.4.1 Effects on Primary Productivity and Carbon Budgeting**

6 Rates of photosynthesis and net primary productivity (NPP) of ecosystems typically
7 correlate with metrics of nitrogen availability (Field and Mooney, 1986; Reich et al., 1997a,
8 1997b; Smith et al., 2002) along with other factors. The addition of nitrogen from an exogenous
9 source will alter the productivity of nitrogen-limited ecosystems. In a meta-analysis that included
10 terrestrial, freshwater, and marine ecosystems, Elser et al. (2007) found that there were similar
11 patterns of nitrogen and phosphorus limitation among ecosystem types. This finding is in
12 contrast with the existing paradigm that nitrogen-limitation dominates in terrestrial and marine
13 ecosystems, and phosphorus-limitation dominates in freshwater ecosystems.

14 It is important to distinguish between effects on primary productivity and effects on
15 carbon sequestration. Nitrogen addition to a given ecosystem may increase primary productivity;
16 however, this does not always translate into greater carbon sequestration because carbon lost
17 from the ecosystem by respiration may offset the carbon gained by production. The following
18 section discusses the mechanisms by which atmospheric nitrogen deposition alters productivity
19 and carbon sequestration in different ecosystems.

20 **6.4.1.1 Terrestrial Ecosystems**

21 **Productivity.** Experimental nitrogen additions to forest ecosystems have elicited positive
22 growth responses in some, but not all, organisms (DeWalle et al., 2006; Elvir et al., 2003;
23 Emmett, 1999; Högberg et al., 2006). A meta-analysis by LeBauer and Treseder (2008) of 126
24 nitrogen addition studies showed that most ecosystems are nitrogen-limited with an average
25 increase of 29% in aboveground growth response to nitrogen. The response ratio was significant
26 within temperate forests, tropical forests, temperate grasslands, tropical grasslands, wetlands, and
27 tundra, but not within deserts (LeBauer and Treseder, 2008).

28 Multiple long-term experiments have demonstrated transient growth increases followed
29 by increased mortality, especially at higher rates of fertilization (Elvir et al., 2003; Högberg et

1 al., 2006; Magill and Aber, 2004; McNulty et al., 2005). Forest growth enhancement can
2 potentially exacerbate other nutrient deficiencies, such as calcium, magnesium, or potassium
3 (K^+). An additional line of evidence comes from the experimental nitrogen removal studies:
4 removal of nitrogen and sulfur from throughfall increased tree growth in Europe (Beier et al.,
5 1995; Boxman et al., 1998).

6 Caspersen et al. (2000) found little evidence for growth enhancement due to nitrogen
7 deposition after evaluating tree growth rates in five states (i.e., Minnesota, Michigan, Virginia,
8 North Carolina, and Florida). Magnani et al. (2007) reported a strong positive correlation
9 between estimated average long-term net ecosystem productivity (Levine et al., 1999) and
10 estimated 1990 nitrogen wet deposition (Holland et al., 2005) for 20 forest stands mostly in
11 Western Europe and the conterminous United States, although there have been critiques of the
12 method and the magnitude of these reported effects (De Schrijver et al., 2008; De Vries et al.,
13 2008; Sutton et al., 2008).

14 Nitrogen deposition can affect the patterns of carbon allocation because most growth
15 occurs above ground. This increases the shoot-to-root ratio, which can be detrimental to the plant
16 because of decreased resistance to environmental stressors, such as drought and windthrow
17 (Braun et al., 2003; Fangmeier et al., 1994; Krupa, 2003; Minnich et al., 1995). Nitrogen
18 saturation also leads to the replacement of slow-growing spruce-fir forest stands by fast-growing
19 deciduous forests that cycle nitrogen more rapidly (McNulty et al., 1996; 2005). In the western
20 United States, atmospheric nitrogen deposition has been shown to cause increased litter
21 accumulation and carbon storage in aboveground woody biomass, which in turn may lead to
22 increased susceptibility to more severe fires (Fenn et al., 2003).

23 **Carbon Sequestration.** Nitrogen addition stimulates plant growth in most ecosystems
24 (LeBauer and Treseder, 2008), which may in turn increase carbon sequestration in plant biomass.
25 On the other hand, maintenance respiration is positively correlated with tissue nitrogen content
26 (Reich et al., 2008) and litter with higher nitrogen content also decomposes faster (Berg and
27 Laskowski, 2006). Therefore, increased leaf nitrogen content under conditions elevated nitrogen
28 deposition may result in higher carbon loss by increasing both autotrophic and heterotrophic
29 respiration. Because of the complexity of interactions between nitrogen and carbon cycling, the
30 effects of nitrogen on carbon budgets (quantified input and output of carbon to the ecosystem)
31 are variable.

1 Many nitrogen fertilization studies have investigated the effect of nitrogen addition on
2 ecosystem carbon sequestration. Adams et al. (2005) examined whether nitrogen fertilization
3 affects carbon sequestration of four Douglas-fir plantation sites in the Pacific Northwest. Those
4 sites were initially established as part of the Regional Forest Nutrition Research Project
5 (RFNRP) and received either three or four additions of 224 kilograms (kg) nitrogen(N)/hectare
6 (ha) as urea (672 to 896 kg N/ha total) over 16 years. They found that the nitrogen-fertilized sites
7 (161 megagrams [Mg] C/ha) had an average of 20% more carbon in the aboveground tree
8 biomass compared to unfertilized sites (135 Mg C/ha), and nitrogen-fertilized soils (260 Mg
9 C/ha) had 48% more soil carbon compared to unfertilized soils (175 Mg C/ha). Canary et al.
10 (2000) studied carbon sequestration of another three RFNRP sites. They also found that nitrogen
11 fertilization, a total of 896 to 1120 kg N/ha over a 16-year-period, increased carbon sequestration
12 of Douglas-fir plantations. However, the response magnitudes were smaller than those reported
13 by Adams et al. (2005). Nitrogen fertilization increased tree biomass carbon by 19% (200.2 Mg
14 C/ha for control, and 238.6 Mg C/ha for nitrogen fertilized site) and soil carbon by 6.2% (123
15 Mg C/ha for control, and 131 Mg C/ha for nitrogen fertilized site).

16 In the ISA (U.S. EPA, 2008, Section 3.3.3.1), a meta-analysis was conducted of 17
17 observations from nine studies in U.S. forests to examine the effect of nitrogen fertilization on
18 forest ecosystem carbon content (EC). In that study, EC was defined as the sum of carbon
19 content of vegetation, forest floor, and soil (Johnson et al., 2006). Details on those publications,
20 including study site, ecosystem type, nitrogen addition level, chemical form of nitrogen, and
21 experimental condition, appear in Annex C of the ISA (U.S. EPA, 2008). To avoid possible
22 confounded variability caused by site conditions, this meta-analysis only included studies of
23 those control and treatment sites that experienced the same climatic, soil, and vegetation
24 conditions. The EPA meta-analysis revealed that while there was a great deal of variation in
25 response, overall nitrogen addition, ranging from 25 to 200 kg N/ha/yr, increased EC by 6% for
26 U.S. forest ecosystems. Different from Magnani et al. (2007), this study did not find any
27 correlation between the amount of nitrogen addition and the response magnitudes of ecosystem
28 carbon sequestration.

29 Less is known regarding the effects of nitrogen deposition on carbon budgets of non-
30 forest ecosystems. The EPA meta-analysis, including 16 observations from nine publications,
31 showed that nitrogen addition from 16 to 320 kg N/ha/yr has no significant effect on net

1 ecosystem exchange (NEE) of nonforest ecosystems (U.S. EPA, 2008, Sections 3.3.3.1 and
2 4.3.1.1). Details on those publications, including study site, ecosystem type, nitrogen addition
3 level, chemical form of nitrogen, and experimental condition, are given in Annex C of the ISA
4 (U.S. EPA, 2008).

The evidence is sufficient to infer a causal relationship between nitrogen deposition and the alteration of biogeochemical cycling of carbon in terrestrial ecosystems.

6 **ISA Conclusion.** The ISA (U.S. EPA, 2008, Section
8 4.3.1.1) concluded that the evidence is *sufficient to infer a*
10 *causal relationship* between nitrogen deposition and the
12 alteration of biogeochemical cycling of carbon in terrestrial
13 ecosystems. Nitrogen is often the most limiting nutrient to growth in ecosystems. Nitrogen
14 deposition thus often increases primary productivity; thereby altering the biogeochemical cycling
15 of carbon. Nitrogen deposition can cause changes in ecosystem carbon budgets. However,
16 whether nitrogen deposition increases or decreases, ecosystem carbon-sequestration remains
17 unclear. The meta-analysis conducted for the ISA indicated that nitrogen addition, ranging from
18 25 to 200 kg N/ha/yr, slightly increased ecosystem carbon content in forest ecosystem. However,
19 nitrogen addition, ranging from 16 to 320 kg N/ha/yr, had no significant effect on NEE for
20 nonforest ecosystems.

21 In terrestrial ecosystems, nitrogen deposition can accelerate plant growth and change
22 carbon allocation patterns (e.g. shoot-to-root ratio), which can increase susceptibility to severe
23 fires, drought, and wind damage. These effects have been studied in the western United States
24 and Europe (Adams et. al., 2005; Braun et. al., 2003; Canary et. al., 2000; Fenn et. al., 2003).
25 The alteration of primary productivity can also alter competitive interactions among plant
26 species. The increase in growth is greater for some species than for others, leading to possible
27 shifts in population dynamics, species composition, community structure, and, in a few
28 instances, ecosystem type.

29 **6.4.1.2 Wetland Ecosystems**

30 **Productivity.** The 1993 NO_x AQCD (U.S. EPA, 1993) reported that nitrogen
31 applications, ranging from 7 to 3120 kg N/ha/yr, stimulated standing biomass production in
32 wetlands by 6% to 413%. The magnitude of the changes in primary production depended on soil
33 nitrogen availability and limitation of other nutrients. However, negative growth rates were
34 observed for some wetland species that were adapted to low-nitrogen environments. For

1 example, increasing nitrogen availability reduced population growth of *Sarracenia purpurea*
2 (commonly known as the Purple pitcher plant or Side-saddle flower). Gotelli and Ellison (2002)
3 reported that the extinction risk of *S. purpurea* within the next 100 years increased substantially
4 if nitrogen deposition rate increased (1% to 4.7%) from the rate of 4.5 to 6.8 kg N/ha/yr. A study
5 of *Sphagnum fuscum* (Rusty peat moss) in six Canadian peatlands showed a weak, although
6 significant, negative correlation between NPP and nitrogen deposition when deposition levels
7 were greater than 3 kg N/ha/yr ($y = 150 - 3.4(x)$; $r^2=0.01$, $p = 0.04$) (Vitt et al., 2003).

9 **Carbon Sequestration.** In the ISA (U.S. EPA, 2008,
11 Sections 4.3.1.1 and 4.3.2.1), a meta-analysis was conducted
13 that included wetlands with other nonforest ecosystems, and
15 the results indicated *no effect* of nitrogen deposition on
17 overall NEE of carbon. In other words, any gain in carbon
18 capture by photosynthesis was offset by ecosystem respiration and carbon leaching. There were
19 not enough studies to evaluate wetlands as a separate category. A study of 23 ombrotrophic
20 peatlands in Canada with deposition levels ranging from 2.7 to 8.1 kg N/ha/yr showed that peat
21 accumulation increases linearly with nitrogen deposition; however, in recent years this rate has
22 begun to slow, indicating limited capacity for nitrogen to stimulate accumulation (Turunen et al.,
23 2004). Soil respiration has been studied in European countries under a natural gradient of
24 atmospheric nitrogen deposition from 2 to 20 kg N/ha/yr. It was found that enhanced
25 decomposition rates for material accumulated under higher atmospheric nitrogen supplies
26 resulted in higher CO₂ emissions and dissolved organic carbon release (Bragazza et al. 2006).

The evidence is sufficient to infer a causal relationship between nitrogen deposition and the alteration of biogeochemical cycling of carbon in transitional ecosystems.

27 **ISA Conclusion.** The ISA (U.S. EPA, 2008, Section 4.3.2.1) concluded that the evidence
28 is *sufficient to infer a causal relationship* between nitrogen deposition and the alteration of
29 biogeochemical cycling of carbon in transitional ecosystems. Nitrogen deposition often increases
30 ecosystem productivity of wetlands, but it also leads to negative population growth rates of some
31 wetland species that were adapted to low-nitrogen environments.

32 There was little evidence for an apparent effect on ecosystem carbon sequestration on
33 wetlands.

6.4.1.3 Aquatic Ecosystems

Productivity. In a meta-analysis of more than 600 experiments, Elser et al. (2007) found that nitrogen limitation occurs frequently in freshwater ecosystems, in contrast to the traditional paradigm that freshwater ecosystems are mainly phosphorus-limited. Numerous other studies have also provided strong evidence indicating that nitrogen deposition has played an important role in influencing the productivity of oligotrophic, high-elevation lakes in the western United States and Canada, as well as in the Canadian Arctic (Das et al., 2005; Lafrancois et al., 2003; Saros et al., 2005; Wolfe et al., 2001, 2003, 2006). A comprehensive study of available data from the northern hemisphere surveys of lakes along gradients of nitrogen deposition showed increased inorganic nitrogen concentrations and productivity to be correlated with atmospheric nitrogen deposition (Bergström and Jansson, 2006).

Estuaries and coastal waters tend to be nitrogen-limited and are, therefore, inherently sensitive to increased atmospheric nitrogen loading (D'Elia et al., 1986; Elser et al., 2007; Howarth and Marino, 2006). There is a strong scientific consensus that nitrogen is the principal cause of coastal eutrophication in the United States (see the Aquatic Nutrient Enrichment Case Study in Chapter 5 and Appendix 6 of this Risk and Exposure Assessment; NRC, 2000).

Carbon Sequestration. Little information is reported regarding the effects of nitrogen deposition on carbon budgets of freshwater, estuarine, and near coastal ecosystems.

ISA Conclusion. The ISA (U.S. EPA, 2008, Section 4.3.3) concluded that the evidence is *sufficient to infer a causal relationship* between nitrogen deposition and the alteration of biogeochemical cycling of carbon in aquatic ecosystems. The productivity of many freshwater ecosystems is nitrogen-limited. Nitrogen deposition can alter species assemblages and cause eutrophication of aquatic ecosystems where nitrogen is the growth-limiting nutrient. In estuarine ecosystems, nitrogen from atmospheric and nonatmospheric sources contributes to increased phytoplankton and algal productivity, leading to eutrophication.

The evidence is sufficient to infer a causal relationship between nitrogen deposition and the alteration of biogeochemical cycling of carbon in aquatic ecosystems.

Ecosystem carbon sequestration is determined by the difference between input (net carbon fixed by photosynthesis) and output (autotrophic and heterotrophic respiration). Although many studies have shown nitrogen increases productivity in aquatic ecosystems, there is a limited understanding on how nitrogen affects NEE or ecosystem respiration of aquatic

1 ecosystems. Quantification of how nitrogen deposition increases or decreases carbon
2 sequestration of freshwater, estuarine, and near-coastal ecosystems remains unclear.

3 **6.4.2 Biogenic Emissions of Nitrous Oxide**

4 **6.4.2.1 Science Overview**

5 Biogenic sources are the dominant contributors (>90%) to atmospheric N₂O. Terrestrial
6 soil is the largest source of atmospheric N₂O, accounting for 60% of global emissions (IPCC,
7 2001). Nitrous oxide production in soil is mainly governed by microbial nitrification and
8 denitrification (Dalal et al., 2003). The contribution of each process to total N₂O production
9 varies with environmental conditions. Denitrifying bacteria reduce NO₃⁻ or nitrite (NO₂⁻) into
10 N₂O or nitrogen (N₂) under anaerobic conditions. In submerged soils, such as wetland soil,
11 denitrification should be the dominant process to N₂O emissions (Conrad, 1996). Increasing
12 NO₃⁻ input generally increases the denitrification rate under suitable conditions of temperature
13 and organic carbon supply. High soil NO₃⁻ concentrations also inhibit the reduction of N₂O to N₂
14 and result in a high N₂O/N₂ ratio (Dalal et al., 2003). Under aerobic environments, autotrophic
15 nitrifying bacteria obtain energy by reducing ammonium (NH₄⁺). Nitrous oxide is an
16 intermediate product of the oxidation of NH₄⁺ to NO₂⁻ or the decomposition of NO₂⁻. The
17 increase in N₂O emissions following NH₄⁺ addition has been observed in many laboratory and
18 field experiments (Aerts and De Caluwe, 1999; Aerts and Toet, 1997; Keller et al., 2005).

19 EPA conducted a meta-analysis, including 99 observations from 30 publications, to
20 evaluate the effects of nitrogen addition on N₂O emissions from nonagricultural ecosystems
21 (U.S. EPA, 2008, Section 3.3.4.2). Details on those publications, including study site, ecosystem
22 type, nitrogen addition level, chemical form of nitrogen, and experimental condition appear in
23 Annex C of the ISA (U.S. EPA, 2008). Overall, the results of the meta-analysis indicated that
24 nitrogen addition, ranging from 10 to 562 kg N/ha/yr, significantly increased N₂O emissions by
25 230% across all ecosystems. Ecosystem type, chemical form of nitrogen, and nitrogen addition
26 level affected the response magnitude of N₂O emissions. Compared to other ecosystems, tropical
27 forests emitted more N₂O under nitrogen enrichment condition (+735%). However, this
28 difference was only significant between tropical and coniferous forests. NO₃⁻ caused a higher
29 stimulation (+494%) of N₂O emissions than NH₄⁺ did (+95%). Although the mean response ratio

1 increased with the amount of nitrogen addition, the differences among the three levels (<75, 75–
2 150, and >150 kg N/ha/yr) were not significant.

3 There were no clear dose-response relationships between GHG emission/uptake and the
4 amount of nitrogen addition to nonagricultural ecosystems, a result consistent with observations
5 in agricultural ecosystems (FAO/IFA, 2001). However, Butterbach-Bahl et al. (1998) found that
6 increasing NH_4^+ wet deposition led to a linear increase in N_2O emissions and a decrease in CH_4
7 oxidation at a red spruce forest site. The dose-response relationship was observed at a small scale
8 characterized by homogenous conditions (such as a specific site), in contrast to the large
9 heterogeneous scale investigated in the EPA meta-analysis. This inconsistency is likely caused
10 because GHG production is influenced by multiple interactions of soil, climate, and vegetation
11 (IPCC, 2001).

12 **6.4.2.2 ISA Conclusion**

14 The ISA concluded that the reviewed evidence is
16 *sufficient to infer a causal relationship* between total The
18 ISA also concluded that the evidence reviewed was *sufficient*
20 *to infer a causal relationship* between total reactive nitrogen
22 deposition and the alteration of N_2O flux in wetland
23 ecosystems (U.S. EPA, 2008, Section 4.3.2.1). Averaged across 19 observations from wetland
24 studies, the meta-analysis conducted by EPA indicated that nitrogen addition, ranging from 15.4
25 to 300 kg N/ha/yr, increased wetland N_2O production by 207% (U.S. EPA, 2008, Section
26 4.3.2.1).

Evidence is sufficient to infer a causal relationship between total reactive nitrogen deposition and (a) the alteration of biogeochemical flux of N_2O in terrestrial ecosystems and (b) N_2O flux in wetland ecosystems.

27 **6.4.3 Methane Emissions and Uptake**

28 **6.4.3.1 Science Overview**

29 Atmospheric methane (CH_4) originates mainly (70% to 80%) from biogenic sources (Le
30 Mer and Roger, 2001). Methane is produced in an anaerobic environment by methanogenic
31 archaea (a type of single-celled organism) bacteria during decomposition of organic matter. Once
32 produced in soil, CH_4 can then be released to the atmosphere or oxidized by methanotrophic
33 bacteria in the aerobic zone (Le Mer and Roger, 2001). Methane production and oxidation
34 processes occur simultaneously in most ecosystems. Wetland soils are generally CH_4 sources,

1 accounting for about 20% of global CH₄ emissions. Nonflooded upland soils are the most
2 important biological sink for CH₄, consuming about 6% of the atmospheric CH₄ (Le Mer and
3 Roger, 2001). Numerous studies have demonstrated that nitrogen is an important regulatory
4 factor for both CH₄ production and oxidation (Bodelier and Laanbroek, 2004).

5 The EPA conducted a meta-analysis, including 61 observations from 27 publications, to
6 evaluate the relationship between nitrogen addition and CH₄ flux. Details on those publications,
7 including study site, ecosystem type, nitrogen addition level, chemical form of nitrogen, and
8 experimental condition, appear in Annex C of the ISA (U.S. EPA, 2008). The impacts of
9 nitrogen addition on CH₄ source and sink strength were estimated by CH₄ emissions and CH₄
10 uptake, respectively.

11 Nitrogen addition, ranging from 30 to 240 kg N/ha/yr significantly increased CH₄
12 emissions by 115% when averaged across all ecosystems. Methane uptake was significantly
14 reduced by 38% under nitrogen addition, ranging from 10 to 560 kg N/ha/yr. Methane uptake
16 was reduced for all ecosystems, but this inhibition was
18 significant only for coniferous and deciduous forest, with a
20 reduction of 28% and 45%, respectively.

The evidence is sufficient to infer a causal relationship between nitrogen deposition and the alteration of biogeochemical flux of CH₄ in terrestrial ecosystems and the alteration of CH₄ flux in wetland ecosystems.

22 Several studies found that CH₄ uptake rates
24 decreased with increasing nitrogen input (Butterbach-Bahl
25 et al., 1998; King and Schnell, 1998; Schnell and King, 1994). However, this meta-analysis did
26 not find significant correlation between the amount of nitrogen addition and the response ratio of
27 CH₄ uptake/emission. The lack of a dose-response relationship likely occurred because CH₄
28 production is influenced by multiple interactions of soil nitrogen content, soil moisture, pH, and
29 temperature (Le Mer and Roger, 2001), and varies greatly over small spatial and temporal scales
30 (IPCC, 2007).

31 **6.4.3.2 ISA Conclusion**

32 The ISA (U.S. EPA, 2008, Section 4.3.1.1) concluded that the evidence is *sufficient to*
33 *infer a causal relationship* between nitrogen deposition and the alteration of biogeochemical flux
34 of CH₄ in terrestrial ecosystems. Averaged across 41 observations from terrestrial ecosystems,
35 including four forms of nitrogen (NH₄⁺, NO₃⁻, NH₄NO₃, and urea) and the addition rates, ranging
36 from 10 to 560 kg N/ha/yr, the meta-analysis conducted by EPA indicated that nitrogen addition

1 reduced CH₄ uptake, but this inhibition was significant only for coniferous and deciduous forests
2 (U.S. EPA, 2008, Section 4.3.1.1).

3 The ISA (U.S. EPA, 2008, Section 4.3.2.1) also concluded that the evidence is *sufficient*
4 *to infer a causal relationship* between nitrogen deposition and the alteration of CH₄ flux in
5 wetland ecosystems. Wetlands are generally net sources of CH₄, but some wetlands can be net
6 sinks, depending on environmental conditions such as drainage and vegetation (Crill et al., 1994;
7 Saarnio et al., 2003). A meta-analysis was performed on a dataset of 17 observations to assess
8 the effects of nitrogen additions on wetland CH₄ fluxes. This dataset included four forms of
9 nitrogen (NH₄⁺, NO₃⁻, NH₄NO₃, and urea) and the addition rates ranged from 30 to 240 kg
10 N/ha/yr. The results indicated that nitrogen addition increased CH₄ production from the wetlands
11 but had no significant effect on CH₄ uptake of wetlands (U.S. EPA, 2008, Section 4.3.2.1).

12 In conclusion, nitrogen addition to ecosystems can affect primary productivity and
13 biogenic GHG fluxes. Due to the complexity of interactions between nitrogen and carbon
14 cycling, the effects of nitrogen on carbon budgets (i.e., quantified input and output of carbon to
15 the ecosystem) are variable. Nitrogen deposition can affect the patterns of carbon allocation
16 because most growth occurs aboveground, and nitrogen deposition also has been found to alter
17 biogeochemical cycling of carbon in transitional ecosystems, such as wetlands, and in aquatic
18 ecosystems. Causal relationships also exists between total reactive nitrogen deposition and (a)
19 the alteration of biogeochemical flux of N₂O in terrestrial and wetland ecosystems and (b) the
20 alteration of biogeochemical flux of CH₄ in terrestrial and wetland ecosystems.

21 **6.4.4 Emission Factors**

22 By adapting the methodology of the Intergovernmental Panel on Climate Change (IPCC)
23 guidelines (Mosier et al., 1998), a nitrogen addition-induced GHG emission/uptake factor (F)
24 can be estimated by the following equation:

$$25 \quad F = (G_N - G_C) / N \quad (2)$$

26 where

27 G_N is annual flux of GHG from fertilized treatment (kg carbon or kg N/ha/yr)

28 G_C is annual flux of GHG from control (kg carbon or kg N/ha/yr)

29 N is annual nitrogen input (kg N/ha/yr).

1 Using the database developed for the EPA EC, N₂O, and CH₄ meta-analyses
2 emission/uptake factors were calculated for the three GHGs. Only field studies that measured
3 growing season or annual GHG fluxes were included in that calculation. Averaged across
4 nitrogen addition treatments ranging from 25 to 200 kg N/ha/yr, the estimated carbon uptake
5 factor (same as C:N response ratio) is 24.5 kg CO₂-C/ha/yr per 1 kg N/ha/yr added to forest
6 ecosystem (n=14), which is much lower than a C:N response of 175 to 225:1 reported by
7 Magnani et al. (2008), but close to the C:N response ratio of 40:1 reported by Högberg (2007)
8 and the C:N response ratio of 50 to 75:1 reported by Sutton et al. (2008). Averaged across
9 nitrogen addition treatments ranging from 10 to 450 kg N/ha/yr, the mean N₂O emissions
10 increased by 0.0087 ± 0.0025 kg N₂O-N/ha/yr per 1 kg N/ha/yr added to the natural ecosystem
11 (n=42), which is comparable to the default N₂O emission factor of 0.0125 kg N₂O-N/ha/yr for
12 agricultural field given by IPCC (2000). Averaged across nitrogen addition treatments ranging
13 from 10 to 450 kg N/ha/yr, the mean CH₄ uptake decreased by 0.015 ± 0.004 kg CH₄-C/ha/yr per
14 1 kg N/ha/yr added to the ecosystem (n=23). There are no emission factors published for which
15 to compare this number. The emission factor for CH₄ was not calculated because there were few
16 field studies that investigated growing season or annual CH₄ emissions under nitrogen addition.

17 **6.4.5 Uncertainty**

18 There is substantial evidence that nitrogen addition causes altered rates of biogenic GHG
19 flux. However, there are limitations to the application of these data to calculate nitrogen
20 deposition effects on net GHG fluxes for the United States. The first obstacle is that ecosystems
21 are heterogeneous across the United States and clear dose-response curves are not available for
22 large heterogeneous landscapes. Micrometeorological factors, including temperature, soil
23 moisture, and precipitation, can vary substantially between ecosystems across the large spatial
24 area of the United States. These factors influence the microbial response to nitrogen addition and
25 introduce variation into the dose-response relationship. Another way to evaluate nitrogen effect
26 on GHG flux is by emission factors (see Section 6.5 of this chapter). Emission factors are
27 calculated by combining data from a range of nitrogen addition levels to produce one quantified
28 rate. This method is a coarse evaluation that introduces several uncertainties: (1) the range of
29 nitrogen addition by studies that are included if the emission factor exceeds those which would

1 be caused by deposition, and (2) the emission factor does not take into account how shifting
2 micrometeorology causes variation in flux rates.

3 **6.5 DIRECT PHYTOTOXIC EFFECTS OF GASEOUS SO_x AND NO_x**

4 The current secondary NAAQS for SO_x and NO_x were set to protect against direct
5 damage to vegetation by the gaseous forms of these pollutants. Uptake of these gaseous
6 pollutants in a plant canopy is a complex process involving adsorption to surfaces (leaves, stems,
7 and soil) and absorption into leaves. These pollutants penetrate into leaves through to the
8 stomata, although there is evidence for limited pathways via the cuticle. Pollutants must be
9 transported from the bulk air to the leaf boundary layer in order to get to the stomata. The entry
10 of gases into a leaf is dependent upon the physical and chemical processes of gas phase and
11 surfaces as well as the stomatal aperture. The aperture of the stomata is controlled largely by the
12 prevailing environmental conditions, such as humidity, temperature, and light intensity. When
13 the stomata are closed, as occurs under dark or drought conditions, resistance to gas uptake is
14 very high and the plant has a very low degree of susceptibility to injury. In contrast, mosses and
15 lichens do not have a protective cuticle barrier to gaseous pollutants or stomates and are
16 generally more sensitive to gaseous sulfur and nitrogen than vascular plants (U.S. EPA, 2008).

17 Outlined below are the effects of the major SO_x and NO_x gases that have phytotoxic
18 effects on vegetation.

19 **6.5.1 SO₂**

20 Currently, SO₂ is the only criteria pollutant with a secondary NAAQS distinct from the
21 primary standard. This standard is intended to protect acute foliar injury resulting from SO₂
22 exposure. The standard is a 3-hour average of 0.50 ppm and was promulgated in 1970 to protect
23 against acute foliar injury in vegetation. The last AQCD for ecological effects of SO_x was
24 completed in 1982 and concluded that controlled experiments and field observations supported
25 retaining this secondary standard (U.S. EPA, 1971, 1982a, 1982c).

26 Acute foliar injury usually happens with hours of exposure, involves a rapid absorption of
27 a toxic dose, and involves collapse or necrosis of plant tissues. Another type of visible injury is
28 termed chronic injury and is usually a result of variable SO₂ exposures over the growing season.
29 The appearance of foliar injury can vary significantly between species and growth conditions

1 affecting stomatal conductance. Currently, no regular monitoring occurs for SO₂ foliar injury
2 effects in the United States.

3 Besides foliar injury, long-term lower SO₂ concentrations can result in reduced
4 photosynthesis, growth, and yield of plants. These effects are cumulative over the season and are
5 often not associated with visible foliar injury. As with foliar injury, these effects vary among
6 species and growing environment. SO₂ is also considered to be the primary factor causing the
7 death of lichens in many urban and industrial areas, with fruticose lichens being more susceptible
8 to SO₂ than many foliose and crustose species (Hutchinson et al., 1996). Damage caused to
9 lichens in response to SO₂ exposure includes reduced photosynthesis and respiration, damage to
10 the algal component of the lichen, leakage of electrolytes, inhibition of nitrogen fixation, reduced
11 K⁺ absorption, and structural changes (Belnap et al., 1993; Farmer et al., 1992; Hutchinson et al.,
12 1996). The 1982 SO_x AQCD summarized the concentration-response information available at the
13 time (U.S. EPA, 1982b). Effects on growth and yield of vegetation were associated with
14 increased SO₂ exposure concentration and time of exposure. However, that document concluded
15 that more definitive concentration-response studies were needed before useable exposure metrics
16 could be identified.

17 Because of falling ambient SO₂ concentrations and focus on O₃ vegetation effects
18 research, few studies have emerged to better inform a metric and levels of concern for effects of
19 SO₂ on growth and productivity of vegetation. The few new studies published since the 1982
20 SO_x AQCD continue to report associations between exposure to SO₂ and reduced vegetation
21 growth. However, the majority of these studies have been performed outside the United States
22 and at levels well above ambient concentrations observed in the United States. In light of limited
23 new data, there is little evidence of phytotoxic effects on vegetation below the level of the
24 current standard. However, the current evidence to date supports the appropriateness of the
25 current standard level to protect vegetation from phytotoxic effects at higher exposure levels.

26 **6.5.2 NO, NO₂ and Peroxyacetyl Nitrate (PAN)**

27 It is well known that in sufficient concentrations, NO, NO₂, and PAN can have phytotoxic
28 effects on plants through decreasing photosynthesis and induction of visible foliar injury
29 (U.S. EPA, 1993). However, the 1993 NO_x AQCD concluded that concentrations of NO, NO₂,
30 and PAN in the atmosphere are rarely high enough to have phytotoxic effects on vegetation

1 (U.S. EPA, 1993). The current ISA (U.S. EPA, 2008, Section 3.4) stated that very little new
2 research has been done on these phytotoxic effects at concentrations currently observed in the
3 United States.

4 The functional relationship between ambient concentrations of NO or NO₂ and a specific
5 plant response, such as foliar injury or growth, is complex. Factors such as inherent rates of
6 stomatal conductance and detoxification mechanisms and external factors, including plant water
7 status, light, temperature, humidity, and the particular pollutant exposure regime, all affect the
8 amount of a pollutant needed to cause symptoms of foliar injury. Plant age and growing
9 conditions, and experimental exposure techniques also vary widely among reports of
10 experimental exposures of plants to NO₂. An analysis conducted in the 1993 NO_x AQCD of over
11 50 peer-reviewed reports on the effects of NO₂ on foliar injury indicated that plants are relatively
12 resistant to NO₂, especially in comparison to foliar injury caused by exposure to O₃ (U.S. EPA,
13 1993). With few exceptions, visible injury was not reported at concentrations below 0.20 ppm,
14 and these occurred when the cumulative duration of exposures extended to 100 hours or longer.
15 Reductions in rates of photosynthesis have also been recorded in experimental exposures of
16 plants to both NO and NO₂, but usually at concentrations significantly higher than would
17 normally be encountered in ambient air (U.S. EPA, 2008).

18 Since the 1993 NO_x AQCD was completed, the current ISA (U.S. EPA, 2008) found
19 most new research on NO₂ exposure to vegetation has taken place in Europe and other areas
20 outside the United States. For example, foliar nitrate (NO₃⁻) reductase activity was increased in
21 Norway spruce (*Picea abies*) trees growing near a highway with average exposures of about
22 0.027 ppm compared to trees growing 1,300 meters away from the highway with NO₂ exposures
23 less than 0.005 ppm (Ammann et al., 1995). This was consistent with other studies on Norway
24 spruce in the field and laboratory (Von Ballmoos et al. 1993; Thoene et al., 1991). Muller et al.
25 (1996) found that the uptake rate of NO₃⁻ by roots of Norway spruce seedlings was decreased by
26 the exposure to 0.1 ppm of NO₂ for 48 hours. Similarly, soybean plants grown in Australia had
27 decreased NO₃⁻ uptake by roots and reduced growth of plants exposed to 1.1 ppm of NO₂ for 7
28 days (Qiao and Murray, 1998). In a Swiss study, poplar cuttings exposed to 0.1 ppm of NO₂ for
29 approximately 12 weeks resulted in decreased stomatal density and increased specific leaf
30 weight, but did not result in other effects such as leaf injury or a change in growth (Gunthardt-
31 Goerg et al., 1996). However, NO₂ enhanced the negative effects of ozone on these poplars,

1 including leaf injury, when the pollutants were applied in combination (Gunthardt-Goerg et al.,
2 1996).

3 Peroxyacetyl nitrate (PAN) is a well-known photochemical oxidant, often co-occurring
4 with O₃ during high photochemical episodes, and PAN has been shown to cause injury to
5 vegetation (See reviews by Cape, 2003, 1997; Kleindienst, 1994; Smidt, 1994; Temple and
6 Taylor, 1983). Acute foliar injury symptoms resulting from exposure to PAN are generally
7 characterized as a glazing, bronzing, or silvering of the underside of the leaf surface; some
8 sensitive plant species include spinach, Swiss chard, lettuces, and tomatoes. Petunias have also
9 been characterized as sensitive to PAN exposures and have been used as bioindicators of in areas
10 of Japan (Nouchi et al., 1984). Controlled experiments have also shown significant negative
11 effects on the net photosynthesis and growth of petunia (*Petunia hybrida* L.) and kidney bean
12 (*Phaseolus vulgaris* L.) after exposure of 30 ppb of PAN for four hours on each of three alternate
13 days (Izuta et al., 1993). As mentioned previously, it is known that oxides of nitrogen, including
14 PAN, could be altering the nitrogen cycle in some ecosystems, especially in the western United
15 States, and contributing nitrogen saturation (Bytnerowicz and Fenn, 1996; Fenn et al., 2003, see
16 Section 3.3). However, PAN is a very small component of nitrogen deposition in most areas of
17 the United States. Although PAN continues to persist as an important component of
18 photochemical pollutant episodes, there is little evidence in recent years suggesting that PAN
19 poses a significant risk to vegetation in the United States (U.S. EPA, 2008).

20 **6.5.3 Nitric Acid (HNO₃)**

21 Relatively little is known about the direct effects of HNO₃ vapor on vegetation. However,
22 the current ISA (U.S. EPA, 2008) highlighted recent research identifying HNO₃ as the cause for
23 decline of sensitive lichen species in areas with relatively high HNO₃ deposition. Further, HNO₃
24 has a very high deposition velocity compared to other pollutants and may be an important source
25 of nitrogen for plants (Hanson and Lindberg, 1991; Hanson and Garten, 1992; Vose and Swank,
26 1990). This deposition could contribute to nitrogen saturation of some ecosystems near sources
27 of photochemical smog (Fenn et al., 1998). For example, in mixed conifer forests (MCFs) of the
28 Transverse Range (i.e., Los Angeles basin mountain ranges), HNO₃ has been estimated to
29 provide 60 percent of all dry deposited nitrogen (Bytnerowicz et al., 1999).

1 HNO₃ deposition has been suspected as the cause of a dramatic decline in lichen species
2 in the Los Angeles basin (Nash and Sigal, 1999). This suggestion was strengthened by an
3 experiment that transplanted *Ramalina* lichen species from clean air habitats (Mount Palomar
4 and San Nicolas Island) to analogous polluted habitats in the Los Angeles basin and repeatedly
5 observed death of the lichens over a few weeks in the summer (Boonpragob and Nash, 1991).
6 Associated with this death was massive accumulation of H⁺ and NO₃⁻ by the lichen thalli
7 (bodies) (Boonpragob et al., 1989). Recently, Riddell et al. (2008) exposed the healthy *Ramalina*
8 *menziesii* thalli to moderate (8–10 ppb) and high (10–14 ppb) HNO₃ concentrations in month-
9 long fumigations and reported a significant decline in chlorophyll content and carbon exchange
10 capacity compared to thalli in control chambers. Thalli treated with HNO₃ showed visual signs
11 of bleaching and were clearly damaged and dead by day 28. The damage may have occurred
12 through several mechanisms including acidification of pigments and cell membrane damage
13 (Riddell et al., 2008). The authors concluded that *Ramalina menziesii* has an unequivocally
14 negative response to the HNO₃ concentrations common to ambient summer conditions in the Los
15 Angeles air basin and that it is very likely that HNO₃ has contributed to the disappearance of this
16 sensitive lichen species from the Los Angeles air basin, as well as other locations with arid
17 conditions and high HNO₃ deposition loads (Riddell et al., 2008).

18 At high ambient concentrations, HNO₃ can also cause damage to vascular plants (U.S.
19 EPA, 2008). Seedlings of ponderosa pine and California black oak subjected to short-term
20 exposures from 50–250 ppb of HNO₃ vapor for 12 hours showed deterioration of the pine needle
21 cuticle in light at 50 ppb (Bytnerowicz et al., 1998a). Oak leaves appeared to be more resistant to
22 HNO₃ vapor, however, with 12-hour exposures in the dark at 200 ppb producing damage to the
23 epicuticular wax structure (Bytnerowicz et al., 1998a). The observed changes in wax chemistry
24 caused by HNO₃ and accompanying injury to the leaf cuticle (Bytnerowicz et al., 1998a) may
25 predispose plants to damage by various environmental stresses such as drought, pathogens, and
26 other air pollutants. Because elevated concentrations of HNO₃ and ozone co-occur in
27 photochemical smog (Solomon et al. 1988), synergistic interactions between the two pollutants
28 are possible (Bytnerowicz et al., 1998b). It should be noted that the experiments described above
29 were observed at relatively short-term exposures at above ambient concentrations of HNO₃.
30 Long-term effects of lower air concentrations that better approximate ambient HNO₃
31 concentrations should be investigated.

1 6.6 REFERENCES

- 2 Adams, A.B., R.B. Harrison, R.S. Sletten, B.D. Strahm, E.C. Turnblom, and C.M. Jensen. 2005.
3 Nitrogen-fertilization impacts on carbon sequestration and flux in managed coastal
4 Douglas-fir stands of the Pacific Northwest. *Forest Ecology and Management* 220:313–
5 325.
- 6 Aerts, R., and H. De Caluwe. 1999. Nitrogen deposition effects on carbon dioxide and methane
7 emissions from temperate peatland soils. *Oikos: A Journal of Ecology* 84:44–54.
- 8 Aerts, R., and S. Toet. 1997. Nutritional controls on carbon dioxide and methane emission from
9 Carex-dominated peat. *Soil Biology and Biochemistry* 29(11–12):1683–1690.
- 10 Back, R.C., J.P. Hurley, and K.R. Rolffhus. 2002. Watershed influences on the transport, fate and
11 bioavailability of mercury in Lake Superior: Field measurements and modeling
12 approaches. *Lakes and Reservoirs: Research and Management* 7:201–206.
- 13 Becker, D.S., and G.N. Bigam. 1995. Distribution of mercury in the aquatic food web of
14 Onondagal Lake, NY. *Water, Air, and Soil Pollution* 80:563–571.
- 15 Beier, C., H. Hultberg, F. Moldan, and R.F. Wright. 1995. MAGIC applied to roof experiments
16 (Risø, Denmark; Gårdsjön, Sweden; Klosterhede, Denmark) to evaluate the rate of reversibility of
17 acidification following experimentally reduced acid deposition. *Water, Air, and Soil*
18 *Pollution* 85:1745–1751.
- 19 Benoit, J.M., C. Gilmour, A. Heyes, R.P. Mason, and C. Miller. 2003. Geochemical and
20 biological controls over methylmercury production and degradation in aquatic
21 ecosystems. Pp. 262–297 in *Biogeochemistry of Environmentally Important Trace*
22 *Elements*. Edited by Y. Chai and O.C. Braids. ACS Symposium Series no. 835.
23 Washington, DC: American Chemical Society.
- 24 Berg, B., and R. Laskowski (eds.). 2006. *Litter Decomposition: A Guide to Carbon and Nutrient*
25 *Turnover*. Advances in Ecological Research, Volume 38. New York: Academic Press.

- 1 Bergström, A.-K., and M. Jansson. 2006. Atmospheric nitrogen deposition has caused nitrogen
2 enrichment and eutrophication of lakes in the northern hemisphere. *Global Change*
3 *Biology* 12:635–643.
- 4 Bloom, N.S. 1992. On the chemical form of mercury in edible fish and marine invertebrate
5 tissue. *Canadian Journal of Fisheries and Aquatic Sciences* 49:1010–1017.
- 6 Bodelier, P.L.E., and H.J. Laanbroek. 2004. Nitrogen as a regulatory factor of methane oxidation
7 in soils and sediments *FEMS Microbiology Ecology* 47(3):265–277.
- 8 Boxman, A.W., K. Blanck, T.-E. Brandrud, B.A. Emmett, P. Gundersen, R.F. Hogervorst, O.J.
9 Kjonass, H. Persson, V. Timmermann. 1998. Vegetation and soil biota response to
10 experimentally-changed nitrogen inputs in coniferous forest ecosystems of the NITREX
11 project. *Forest Ecology and Management* 101:65–79.
- 12 Bragazza, L., C. Freeman, T. Jones, H. Rydin, J. Limpens, N. Fenner, T. Ellis, R. Gerdol, M.
13 Hajek, and T. Hajek. 2006. Atmospheric nitrogen deposition promotes carbon loss from
14 peat bogs. *Proceedings of the National Academy of Sciences of the United States of*
15 *America* 103:19386–19389.
- 16 Branfireun, B.A., N.T. Roulet, C.A. Kelly, and J.W.M. Rudd. 1999. *In situ* sulphate stimulation
17 of mercury methylation in a boreal peatland: toward a link between acid rain and
18 methylmercury contamination in remote environments. *Global Biogeochemical Cycles*
19 *13*:743–750.
- 20 Braun, S., C. Schindler, R. Volz, and W. Flückiger. 2003. Forest damages by the storm ‘Lothar’
21 in permanent observation plots in Switzerland: The significance of soil acidification and
22 nitrogen deposition. *Water, Air, and Soil Pollution* 142:327–340.
- 23 Butterbach-Bahl, K., R. Gasche, C.H. Huber, K. Kreutzer, and H. Papen. 1998. Impact of N-
24 input by wet deposition on N-trace gas fluxes and CH₄-oxidation in spruce forest
25 ecosystems of the temperate zone in Europe. *Atmospheric Environment* 32:559–564.

- 1 Canary, J.D., R.B. Harrison, J.E. Compton, and H.N. Chappell. 2000. Additional carbon
2 sequestration following repeated urea fertilization of second-growth Douglas-fir stands in
3 western Washington. *Forest Ecology and Management* 138:225–232.
- 4 Caspersen, J.P., S.W. Pacala, J.C. Jenkins, G.C. Hurtt, P.R. Moorcroft, and R.A. Birdsey. 2000.
5 Contributions of land-use history to carbon accumulation in U.S. forests. *Science*
6 290:1148–1151.
- 7 Chen, C.Y., R.S. Stemberger, N.C. Kamman, B.M. Mayes, and C.L. Folt. 2005. Patterns of Hg
8 bioaccumulation and transfer in aquatic food webs across multi-lake studies in the
9 Northeast US. *Ecotoxicology* 14(1–2):135–147.
- 10 Compeau, G.C., and R. Bartha. 1985. Sulfate-reducing bacteria: principal methylators of
11 mercury in anoxic estuarine sediment. *Applied and Environmental Microbiology* 50:498–
12 502.
- 13 Conrad, R. 1996. Soil microorganisms as controllers of atmospheric trace gases (H₂, CO, CH₄,
14 OCS, N₂O, and NO). *Microbiology Review* 60(4):609–640.
- 15 Crill, P.M., P.J. Martikainen, H. Nykanen, and J. Silvola. 1994. Temperature and N-fertilization
16 effects on methane oxidation in a drained peatland soil. *Soil Biology and Biochemistry*
17 26:1331–1339.
- 18 D’Elia, C.J., J.G. Sanders, and W.R. Boynton. 1986. Nutrient enrichment studies in a coastal
19 plain estuary: phytoplankton growth in large-scale, continuous cultures. *Canadian*
20 *Journal of Fisheries and Aquatic Sciences* 43:397–406.
- 21 Dalal, R.C., W.J. Wang, G.P. Robertson, and W.J. Parton. 2003. Nitrous oxide emission from
22 Australian agricultural lands and mitigation options: a review. *Australian Journal of Soil*
23 *Research* 41:165–195.
- 24 Das, B., R.D. Vinebrooke, A. Sanchez-Azofeifa, B. Rivard, and A.P. Wolfe. 2005. Inferring
25 sedimentary chlorophyll concentrations with reflectance spectroscopy: a novel approach
26 to reconstructing historical changes in the trophic status of mountain lakes. *Canadian*
27 *Journal of Fisheries and Aquatic Sciences* 62:1067–1078.

- 1 De Schrijver, A., K. Verheyen, J. Mertens, J. Staelens, K. Wuyts, and B. Muys. 2008. Nitrogen
2 saturation and net ecosystem production. (Brief communication arising from: F. Magnani
3 et al. (2007). *Nature* 447:849–850; Magnani et al. reply). *Nature* 451:E1.
- 4 De Vries, W., S. Solberg, M. Dobbertin, H. Sterba, D. Laubhahn, G.J. Reinds, G.J. Nabuurs, P.
5 Gundersen, and M.A. Sutton. 2008. Ecologically implausible carbon response? (Brief
6 communication arising from: F. Magnani et al. (2007). *Nature* 447:849–850; Magnani
7 et al. reply). *Nature* 451:E1–E3.
- 8 DeWalle, D.R., J.N. Kochenderfer, M.B. Adams, G.W. Miller, F.S. Gilliam, F. Wood, S.S.
9 Odenwald-Clemens, and W.E. Sharpe. 2006. Vegetation and acidification. Pp. 137–1888
10 in *The Fernow Watershed Acidification Study*. Edited by M.B. Adams, D.R. DeWalle,
11 and J.L. Hom. Dordrecht, The Netherlands: Springer.
- 12 Drevnick, P.E., D.E. Canfield, P.R. Gorski, A.L.C. Shinneman, D.R. Engstrom, D.C.G. Muir,
13 G.R. Smith, P.J. Garrison, L.B. Cleckner, J.P. Hurley, R.B. Noble, R.R. Otter, and J.T.
14 Oris. 2007. Deposition and cycling of sulfur controls mercury accumulation in Isle
15 Royale fish. *Environmental Science and Technology* 41(21):7266–7272.
- 16 Driscoll, C.T., C. Yan, C.L. Schofield, R. Munson, and J. Holsapple. 1994. The mercury cycle
17 and fish in the Adirondack lakes. *Environmental Science and Technology* 28:136A–
18 143A.
- 19 Driscoll, C. T., G. B. Lawrence, A. J. Bulger, T. J. Butler, C. S. Cronan, C. Eagar, K.
20 F. Lambert, G. E. Likens, J. L. Stoddard, and K. C. Weathers. 2001. Acidic deposition in
21 the northeastern United States: Sources and inputs, ecosystem effects, and management
22 strategies. *BioScience* 51:180–198.
- 23 Driscoll, C.T., Y-J. Han, C.Y. Chen, D.C. Evers, K.F. Lambert, T.M. Holsen, N.C. Kamman,
24 and R.K. Munson. 2007. Mercury contamination in forest and freshwater ecosystems in
25 the northeastern United States. *BioScience* 57(1):17–28.
- 26 Elser, J.J., M.E.S. Bracken, E.E. Cleland, D.S. Gruner, W.S. Harpole, H. Hillebrand II, J.T. Ngai,
27 E.W. Seabloom, J.B. Shurin, and J.E. Smith. 2007. Global analysis of nitrogen and

- 1 phosphorus limitation of primary producers in freshwater, marine, and terrestrial
2 ecosystems. *Ecology Letters* 10(12):1135–1142.
- 3 Elvir, J.A., G.B. Wiersma, A.S. White, and I.J. Fernandez. 2003. Effects of chronic ammonium
4 sulfate treatment on basal area increment in red spruce and sugar maple at the Bear Brook
5 Watershed in Maine. *Canadian Journal of Forest Research* 33:862–869.
- 6 Emmett, B.A., 1999. The impact of nitrogen on forest soils and feedbacks on tree growth. *Water*
7 *Air and Soil Pollution* 116:65–74.
- 8 Evers, D.C., Y. Han, C.T. Driscoll, N.C. Kamman, M.W. Goodale, K.F. Lambert, T.M. Holsen,
9 C.Y. Chen, T.A. Clair, and T. Butler. 2007. Biological mercury hotspots in the
10 northeastern United States and southeastern Canada. *BioScience* 57:29–43.
- 11 Fangmeier, A., A. Hadwiger-Fangmeier, L. Van der Eerden, and H.J. Jager. 1994. Effects of
12 atmospheric ammonia on vegetation: A review. *Environmental Pollution* 86:43–82.
- 13 FAO/IFA (Food and Agriculture Organization of the United Nations/International Fertilizer
14 Industry Association). 2001. *Global Estimates of Gaseous Emissions of NH₃, NO and*
15 *N₂O from Agricultural Land*. Food and Agriculture Organization of the United Nations,
16 Rome, Italy. Available at <ftp://ftp.fao.org/agl/agll/docs/globest.pdf>.
- 17 Faust, S.D., and M.A. Osman. 1981. Chemistry of natural waters. Mercury, arsenic, lead,
18 cadmium, selenium, and chromium in aquatic environments. In *Chemistry of Natural*
19 *Waters*. Ann Arbor, MI: Ann Arbor Science Publishers.
- 20 Fenn, M.E., J.S. Baron, E.B. Allen, H.M. Rueth, K.R. Nydick, L. Geiser, W.D. Bowman, J.O.
21 Sickman, T. Meixner, D.W. Johnson, and P. Neitlich. 2003. Ecological effects of
22 nitrogen deposition in the western United States. *BioScience* 53:404–420.
- 23 Fenn, M.E., M.A. Poth, J.D. Aber, J.S. Baron, B.T. Bormann, D.W. Johnson, A.D. Lemly, S.G.
24 McNulty, D.F. Ryan, and R. Stottlemyer. (1998). Nitrogen excess in North American
25 ecosystems: Predisposing factors, ecosystem responses, and management strategies.
26 *Ecological Applications*, 8: 706–733.

- 1 Field, C.B., and H.A. Mooney. 1986. The photosynthesis-nitrogen relationship in wild plants. In
2 *On the Economy of Plant Form and Function*. Edited by T.J. Givnish. Cambridge, UK:
3 Cambridge University Press.
- 4 Fleming, E.J., E.E. Mack, P.G. Green, and D.C. Nelson. 2006. Mercury methylation from
5 unexpected sources: Molybdate-inhibited freshwater sediments and an iron-reducing
6 bacterium. *Applied Environmental Microbiology* 72:457–464.
- 7 Gilmour, C.C., E.A. Henry, and R. Mitchell. 1992. Sulfate stimulation of mercury methylation in
8 freshwater sediments. *Environmental Science and Technology* 26:2281–2287.
- 9 Gilmour, C.G., G.S. Riedel, M.C. Ederington, J.T. Bell, J.M. Benoit, G.A. Gill, and M.C.
10 Stordal. 1998. Methylmercury concentration and production rates across a trophic
11 gradient in the northern Everglades. *Biogeochemistry* 40:327–345.
- 12 Gotelli, N.J., and A.M. Ellison. 2002. Nitrogen deposition and extinction risk in the northern
13 pitcher plant, *Sarracenia purpurea*. *Ecology* 83:2758–2765.
- 14 Grieb, T.M., C.T. Driscoll, S.P. Gloss, C.L. Schofield, G.L. Bowie, and D.B. Porcella. 1990.
15 Factors affecting mercury accumulation in fish in the upper Michigan peninsula.
16 *Environmental Toxicology and Chemistry* 9:919–930.
- 17 Hammerschmidt, C.R., W. Fitzgerald, C.H. Lamborg, P.H. Balcom, and P.T. Visscher. 2004.
18 Biogeochemistry of methylmercury in sediments of Long Island Sound. *Marine*
19 *Chemistry* 90:31–52.
- 20 Harmon, S.M., J.K. King, J.B. Gladden, and L.A. Newman. 2007. Using sulfate-amended
21 sediment slurry batch reactors to evaluate mercury methylation. *Archives of*
22 *Environmental Contamination and Toxicology* 52:326–333.
- 23 Harris, H.H., I.J. Pickering, and G.N. George. 2003. The chemical form of mercury in fish.
24 *Science* 301:1203.
- 25 Högberg, P. 2007. Environmental science: Nitrogen impacts on forest carbon. *Nature* 447:781–
26 782.

- 1 Högberg, P., H. Fan, M. Quist, D. Binkleys, and C. Oloftamm. 2006. Tree growth and soil
2 acidification in response to 30 years of experimental nitrogen loading on boreal forest.
3 *Global Change Biology* 12:489–499.
- 4 Holland E.A., B.H. Braswell, J. Sulzman, and J.F. Lamarque. 2005. Nitrogen deposition onto the
5 United States and western Europe: Synthesis of observations and models. *Ecological*
6 *Applications* 15:38–57.
- 7 Howarth, R.W., and R. Marino. 2006. Nitrogen as the limiting nutrient for eutrophication in
8 coastal marine ecosystems: evolving views over three decades. *Limnology and*
9 *Oceanography* 51:364–376.
- 10 Hrabik, T.R., and C.J. Watras. 2002. Recent declines in mercury concentration in a freshwater
11 fishery: isolating the effects of de-acidification and decreased atmospheric mercury
12 deposition in Little Rock Lake. *Science of the Total Environment* 297:229–237.
- 13 IPCC (Intergovernmental Panel on Climate Change). 2000. *Good Practice Guidance and*
14 *Uncertainty Management in National Greenhouse Gas Inventories*. New York:
15 Cambridge University Press.
- 16 IPCC (Intergovernmental Panel on Climate Change). 2001. *Climate Change 2001: Synthesis*
17 *Report*. A Contribution of Working Groups I, II, and III to the Third Assessment Report
18 of the Intergovernmental Panel on Climate Change. Edited by R.T. Watson and the Core
19 Writing Team, Cambridge, United Kingdom, and New York: Cambridge University
20 Press.
- 21 IPCC (Intergovernmental Panel on Climate Change). 2007. *Climate Change 2007: The Physical*
22 *Science Basis*. Contribution of Working Group I to the Fourth Assessment Report of the
23 Intergovernmental Panel on Climate Change. Cambridge, United Kingdom: Cambridge
24 University Press. Available at <http://www.ipcc.ch/ipccreports/ar4-wg1.htm> (accessed
25 December 6, 2007).
- 26 Jeremiason, J.D., D.R. Engstrom, E.B. Swain, E.A. Nater, B.M. Johnson, J.E.
27 Almendinger, B.A. Monson, and R.K. Kolka. 2006. Sulfate addition increases

- 1 methylmercury production in an experimental wetland. *Environmental Science and*
2 *Technology* 40:3800–3806.
- 3 Johnson, D.W., A.M. Hoylman, J.T. Ball, and R.F. Walker. 2006. Ponderosa pine responses to
4 elevated CO₂ and nitrogen fertilization. *Biogeochemistry* 77:157–175.
- 5 Kamman, N.C., P.M. Lorey, C.T. Driscoll, R. Estabrook, A. Major, B. Pientka, and E. Glassford.
6 2004. Assessment of mercury in waters, sediments and biota of New Hampshire and
7 Vermont lakes sampled using a geographically randomized design. *Environmental*
8 *Toxicology and Chemistry* 23:1172–1186.
- 9 Keller, M., R. Varner, J. Dias, H. Silva, P. Crill, R.C. Oliveira, Jr., and G.P. Asner. 2005. Soil-
10 atmosphere exchange of nitrous oxide, nitric oxide, methane, and carbon dioxide in
11 logged and undisturbed forest in the Tapajos National Forest, Brazil. *Earth Interactions*
12 9:1–28.
- 13 Kerin, E.J., C.C. Gilmour, E.E. Roden, M.T. Suzuki, J.D. Coates, and R.P. Mason. 2006.
14 Mercury methylation by dissimilatory iron-reducing bacteria. *Applied and Environmental*
15 *Microbiology* 72:7919–21.
- 16 King, G.M., and S. Schnell. 1998. Effects of ammonium and non-ammonium salt additions on
17 methane oxidation by *Methylosinus trichosporium* OB3b and Maine forest soils. *Applied*
18 *and Environmental Microbiology* 64:253–257.
- 19 Krabbenhoft, D.P., J.G. Wiener, W.G. Brumbaugh, M.L. Olson, J.F. DeWild, and T.J. Sabin.
20 1999. *A National Pilot Study of Mercury Contamination of Aquatic Ecosystems Along*
21 *Multiple Gradients*. Pp. 147–160 in U.S. Geological Survey Toxic Substances Hydrology
22 Program, Proceedings of the Technical Meeting, Charleston, SC, March 8–12. Volume 2
23 of 3, Contamination of Hydrologic Systems and Related Ecosystems, Water-Resources
24 Investigation Report 99-4018B.
- 25 Krupa, S.V. 2003. Effects of atmospheric ammonia (NH₃) on terrestrial vegetation: a review.
26 *Environmental Pollution* 124(2):179–221.

- 1 Lafrancois, B.M., K.R. Nydick, and B. Caruso. 2003. Influence of nitrogen on phytoplankton
2 biomass and community composition in fifteen Snowy Range lakes (Wyoming, U.S.A.).
3 *Arctic, Antarctic, and Alpine Research* 35(4):499–508.
- 4 Langer, C.S., W.F. Fitzgerald, P.T. Visscher, and G.M. Vandal. 2001. Biogeochemical cycling of
5 methylmercury at Barn Island salt marsh, Stonington, CT, USA. *Wetlands Ecology and*
6 *Management* 9:295–310.
- 7 LeBauer, D.S., and K.K. Treseder. 2008. Nitrogen limitation of net primary productivity in
8 terrestrial ecosystems is globally distributed. *Ecology* 89:371–379.
- 9 LeMer, J., and P. Roger. 2001. Production, oxidation, emission and consumption of methane by
10 soils: A review. *European Journal of Soil Biology* 37:25–50.
- 11 Levine, J.S., T. Bobbe, N. Ray, and R.G. Witt. 1999. Wildland Fires and the Environment: A
12 Global Synthesis. UNEP/DEIAEW/TR.99-1. United Nations Environmental Programme,
13 Division of Environmental Information, Assessment and Early Warning, Nairobi, Kenya.
14 Available at <http://www.na.unep.net/publications/wildfire.pdf>.
- 15 Magill, A.H., and J.D. Aber. 2004. Ecosystem response to 15 years of chronic nitrogen additions
16 at the Harvard Forest LTER, Massachusetts, USA. *Forest Ecology and Management*
17 *196*:7–28.
- 18 Magnani, F., M. Mencuccini, M. Borghetti, P. Berbigier, F. Berninger, S. Delzon, A. Grelle, P.
19 Hari, P.G. Jarvis, P. Kolari, A.S. Kowalski, H. Lankreijer, B.E. Law, A. Lindroth, D.
20 Loustau, G. Manca, J.B. Moncrieff, M. Rayment, V. Tedeschi, R. Valentini, and J. Grace.
21 2007. The human footprint in the carbon cycle of temperate and boreal forests. *Nature*
22 *447*:848–850.
- 23 Magnani, F., M. Mencuccini, M. Borghetti, F. Berninger, S. Delzon, A. Grelle, P. Hari, P.G.
24 Jarvis, P. Kolari, and A.S. Kowalski. 2008. Magnani et al. reply. *Nature* 451:3.
- 25 Mahrt, L. 1998. Stratified atmospheric boundary layers and breakdown of models. *Theoretical*
26 *and Computational Fluid Dynamics* 11:263–279.

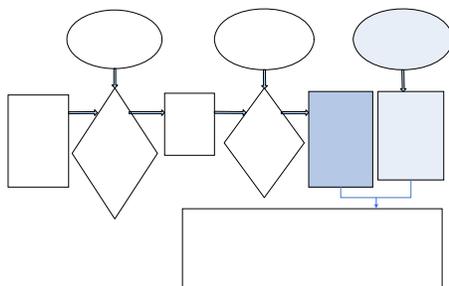
- 1 Mauro, J.B.N., J.R.D. Guimarães, H. Hintelmann, C. Watras, L. Haack, and A.S. Coelho-Souza.
2 2002. Mercury methylation in macrophytes, periphyton and water: Comparative studies
3 with stable and radio-mercury additions. *Analytical and Bioanalytical Chemistry*
4 374:983–989.
- 5 McNulty, S.G., J.D. Aber, and S.D. Newman. 1996. Nitrogen saturation in a high elevation New
6 England spruce-fir stand. *Forest Ecology and Management* 84:109–121.
- 7 McNulty, S.G., J. Boggs, J.D. Aber, L. Rustad, and A. Magill. 2005. Red spruce ecosystem level
8 changes following 14 years of chronic N fertilization. *Forest Ecology and Management*
9 219:279–291.
- 10 Mierle, G., and R. Ingram. 1991. The role of humic substances in the mobilization of mercury
11 from watersheds. *Water, Air, and Soil Pollution* 56:349–357.
- 12 Minnich, R.A., M.G. Barbour, J.H. Burk, and R.F. Fernau. 1995. Sixty years of change in
13 Californian conifer forests of the San Bernardino Mountains. *Conservation Biology*
14 9:902–914.
- 15 Mitchell, C.P.J., B.A. Branfireun, and R.K. Kolka. 2008. Assessing sulfate and carbon controls
16 on net methylmercury production in peatlands: An in situ mesocosm approach. *Applied*
17 *Geochemistry* 23:503–518.
- 18 Mosier, A., C. Kroeze, C. Nevison, O. Oenema, S. Seitzinger, and O. van Cleemput. 1998.
19 Closing the global N₂O budget: Nitrous oxide emissions through the agricultural nitrogen
20 cycle - OECD/IPCC/IEA phase II development of IPCC guidelines for national
21 greenhouse gas inventory methodology. *Nutrient Cycling in Agroecosystems* 52:225–248.
- 22 Munthe, J., R.A. Bodaly, B.A. Branfireun, C.T. Driscoll, C.C. Gilmour, R. Harris, M. Horvat, M.
23 Lucotte, and O. Malm. 2007. Recovery of mercury-contaminated fisheries. *AMBIO:A*
24 *Journal of the Human Environment* 36:33–44.
- 25 Myers, M.D., M.A. Ayers, J.S. Baron, P.R. Beauchemin, K.T. Gallagher, M.B. Goldhaber, D.R.
26 Hutchinson, J.W. LaBaugh, R.G. Sayre, and S.E. Schwarzbach. 2007. USGS goals for
27 the coming decade. *Science* 318:200–201.

- 1 NRC (National Research Council). 2000. *Clean coastal waters: understanding and reducing the*
2 *effects of nutrient pollution*. Washington, DC: National Academy Press.
- 3 Reich, P.B., M.B. Walters, and D.S. Ellsworth. 1997a. From tropics to tundra: Global
4 convergence in plant functioning. *Proceedings of the National Academy of Science*
5 *94*:13730–13734.
- 6 Reich, P.B., D.F. Grigal, J.D. Aber, and S.T. Gower. 1997b. Nitrogen mineralization and
7 productivity in 50 hardwood and conifer stands on diverse soils. *Ecology* *78*:335–347.
- 8 Reich, P.B., M.G. Tjoelker, K.S. Pregitzer, I.J. Wright, J. Oleksyn, and J.L. Machado. 2008.
9 Scaling of respiration to nitrogen in leaves, stems and roots of higher land plants. *Ecology*
10 *Letters* *11*:793–801
- 11 Saarnio, S., S. Jarvio, T. Saarinen, H. Vasander, and J. Silvola. 2003. Minor changes in
12 vegetation and carbon gas balance in a boreal mire under a raised CO₂ or NH₄NO₃
13 supply. *Ecosystems* *6*:46–60.
- 14 Saros, J.E., T.J. Michel, S.J. Interlandi, and A.P. Wolfe. 2005. Resource requirements of
15 *Asterionella formosa* and *Fragilaria crotonensis* in oligotrophic alpine lakes:
16 Implications for recent phytoplankton community reorganizations. *Canadian Journal of*
17 *Fisheries and Aquatic Sciences* *62*:1681–1689.
- 18 Scheuhammer, A.M., and P.J. Blancher. 1994. Potential risk to common loons (*Gavia immer*)
19 from methylmercury exposure in acidified lakes. *Hydrobiologia* *279/280*:445–455.
- 20 Scheuhammer, A.M., M.W. Meyer, M.B. Sandheinrich, and M.W. Murray. 2007. Effects of
21 environmental methylmercury on the health of wild birds, mammals, and fish. *AMBIO: A*
22 *Journal of the human Environment* *36*:12–18.
- 23 Schnell, S., and G.M. King. 1994. Mechanistic analysis of ammonium inhibition of atmospheric
24 methane consumption in forest soils. *Applied and Environmental Microbiology* *60*:3514–
25 3521.

- 1 Smith, M.-L., S.V. Ollinger, M.E. Martin, J.D. Aber, R.A. Hallett, and C.L. Goodale. 2002.
2 Direct estimation of aboveground forest productivity through hyperspectral remote
3 sensing of canopy nitrogen. *Ecological Applications* 12:1286–1302.
- 4 St. Louis, V.L., J.W.M. Rudd, C.A. Kelly, K.G. Beaty, R.J. Flett, and N.T. Roulet. 1996.
5 Production and loss of methylmercury and loss of total mercury from boreal forest
6 catchments containing different types of wetlands. *Environmental Science and*
7 *Technology* 30:2719–2729.
- 8 Suns, K., and G. Hitchin. 1990. Interrelationships between mercury levels in yearling yellow
9 perch, fish condition and water quality. *Water, Air, and Soil Pollution* 50:255–265.
- 10 Sutton, M.A., D. Simpson, P.E. Levy, R.I. Smith, S. Reis, M. van Oijen, and W. de Vries. 2008.
11 Uncertainties in the relationship between atmospheric nitrogen deposition and forest
12 carbon sequestration. *Global Change Biology* 14:2057–2063.
- 13 Turunen, J., N.T. Roulet, T.R. Moore, and P.J.H. Richard. 2004. Nitrogen deposition and
14 increased carbon accumulation in ombrotrophic peatlands in eastern Canada. *Global*
15 *Biogeochemical Cycles* 18.
- 16 U.S. EPA (Environmental Protection Agency). 1971. *Air Quality Criteria for Nitrogen Oxides*.
17 No. AP-84. U.S. Environmental Protection Agency, Air Pollution Control Office,
18 Washington, DC. January.
- 19 U.S. EPA (Environmental Protection Agency) 1982a. *Air Quality Criteria for Oxides of*
20 *Nitrogen*. EPA-600/8-82-026. U.S. Environmental Protection Agency, Office of Research
21 and Development, National Center for Environmental Assessment, Washington, DC.
- 22 U.S. EPA (Environmental Protection Agency). 1982b. *Air Quality Criteria for Particulates and*
23 *Sulfur Oxides*. Volume 1. EPA-600/8-82-029a. U.S. Environmental Protection Agency,
24 Office of Air Quality Planning and Standards, Research Triangle Park, NC. December.
- 25 U.S. EPA (Environmental Protection Agency). 1993. *Air Quality Criteria for Oxides of*
26 *Nitrogen*. EPA/600/8-91/049aF-cF. 3v. U.S. Environmental Protection Agency, Office of

- 1 Health and Environmental Assessment, Environmental Criteria and Assessment Office,
2 Research Triangle Park, NC.
- 3 U.S. EPA (Environmental Protection Agency). 1996. *Air Quality Criteria for Ozone and Related*
4 *Photochemical Oxidants*. EPA/600/AP-93/004aF-cF. 3v. U.S. Environmental Protection
5 Agency, Office of Research and Development, Research Triangle Park, NC. Available at
6 <http://cfpub2.epa.gov/ncea>.
- 7 U.S. EPA (Environmental Protection Agency). 2007a. *National Listing of Fish Advisories*
8 *Technical Fact Sheet: 2005/06 National Listing Fact Sheet*. EPA-823-F-07-003. U.S.
9 Environmental Protection Agency, Office of Water, Washington, DC. Available at
10 <http://www.epa.gov/waterscience/fish/advisories/2006/tech.html>.
- 11 U.S. EPA (Environmental Protection Agency). 2007b. *The U.S. Inventory of Greenhouse Gas*
12 *Emissions and Sinks: Fast Facts*. EPA 430-F-07-004. U.S. Environmental Protection
13 Agency, Office of Atmospheric Programs, Washington, DC. April.
- 14 U.S. EPA (Environmental Protection Agency). 2008. *Integrated Science Assessment (ISA) for*
15 *Oxides of Nitrogen and Sulfur–Ecological Criteria (Final Report)*. EPA/600/R-
16 08/082F. U.S. Environmental Protection Agency, National Center for Environmental
17 Assessment–RTP Division, Office of Research and Development, Research Triangle
18 Park, NC. Available at <http://cfpub.epa.gov/ncea/cfm/recordisplay.cfm?deid=201485>.
- 19 USGS (U.S. Geological Survey). 2006. *Investigations and Monitoring of Mercury in Indiana by*
20 *the U.S. Geological Survey*. U.S. Department of the Interior, U.S. Geological Survey,
21 Indiana Water Science Center, Indianapolis, IN. Available at
22 <http://in.water.usgs.gov/mercury>.
- 23 Vitt, D.H., K. Wieder, L.A. Halsey, and M. Turetsky. 2003. Response of *Sphagnum fuscum* to
24 nitrogen deposition: A case study of ombrogenous peatlands in Alberta, Canada.
25 *Bryologist* 106:235–245.

- 1 Watras, C.J., and K.A. Morrison. 2008. The response of two remote, temperate lakes to changes
2 in atmospheric mercury deposition, sulfate, and the water cycle. *Canadian Journal of*
3 *Fisheries and Aquatic Sciences* 65:100–116.
- 4 Watras, C.J., K.A. Morrison, J.S. Host, and N.S. Bloom. 1995. Concentration of mercury species
5 in relationship to other site-specific factors in the surface waters of northern Wisconsin
6 lakes. *Limnology and Oceanography* 40:556–565.
- 7 Wiener, J.G., D.P. Krabbenhoft, G.H. Heinz, and A.M. Scheuhammer. 2003. Ecotoxicology of
8 mercury. Pp. 409–643 in *Handbook of Ecotoxicology*. 2nd edition. Edited by D.J.
9 Hoffman, B.A. Rattner, G.A. Burton, and J. Cairns. Boca Raton, FL: CRC Press.
- 10 Wolfe, A.P., J.S. Baron, and R.J. Cornett. 2001. Anthropogenic nitrogen deposition induces
11 rapid ecological changes in alpine lakes of the Colorado Front Range (USA). *Journal of*
12 *Paleolimnology* 25:1–7.
- 13 Wolfe, A.P., A.C. Van Gorpe, and J.S. Baron. 2003. Recent ecological and biogeochemical
14 changes in alpine lakes of Rocky Mountain National Park (Colorado, USA): A response
15 to anthropogenic nitrogen deposition. *Geobiology* 1:153–168.
- 16 Wolfe, A.P., C.A. Cooke, and W.O. Hobbs. 2006. Are current rates of atmospheric nitrogen
17 deposition influencing lakes in the eastern Canadian Arctic? *Arctic, Antarctic, and Alpine*
18 *Research* 38:465–476.
- 19



7.0 SYNTHESIS AND INTEGRATION OF CASE STUDY RESULTS

The most recent secondary National Ambient Air Quality Standards (NAAQS) reviews have characterized known or anticipated adverse effects to public welfare by assessing changes in ecosystem structure or processes using a weight-of-evidence approach that uses both quantitative and qualitative data. For example, the previous ozone review evaluated changes in foliar injury, growth loss, and biomass reduction on trees beyond the seedling stage using field measurement data. The presence or absence of foliar damage in counties meeting the current standard has been used as a way to evaluate the impact of current ozone air quality on plants.

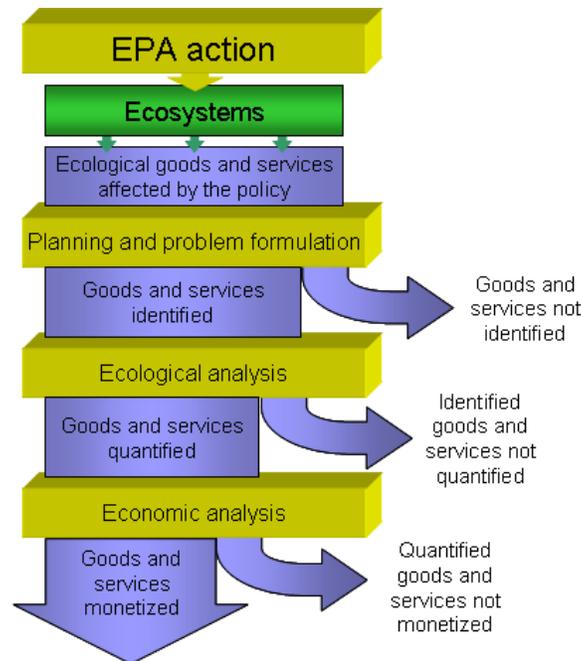
Characterizing a known or anticipated adverse effect to public welfare is an important component of developing any secondary NAAQS. According to the Clean Air Act (CAA), welfare effects include

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 (Section 302(h)).

In other words, welfare effects are those effects that are important to people or that society views as beneficial. A similar concept used by the scientific community is ecosystem services. Ecosystem services can be generally defined as the benefits that individuals and organizations obtain from ecosystems. EPA has defined ecological goods and services 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” (U.S. EPA, 2006). Ecosystem services can be classified as provisioning

1 (e.g., food, water), regulating (e.g., control of climate and disease), cultural (e.g., recreational),
 2 and supporting (e.g., nutrient cycling) (MEA, 2005). Conceptually, changes in ecosystem
 3 services may be used to aid in characterizing a known or anticipated adverse effect to public
 4 welfare. In the context of this review, ecosystem services may also aid in assessing the
 5 magnitude and significance of a resource and in assessing how nitrogen oxides (NO_x) and sulfur
 6 oxides (SO_x) concentrations and deposition may impact that resource.

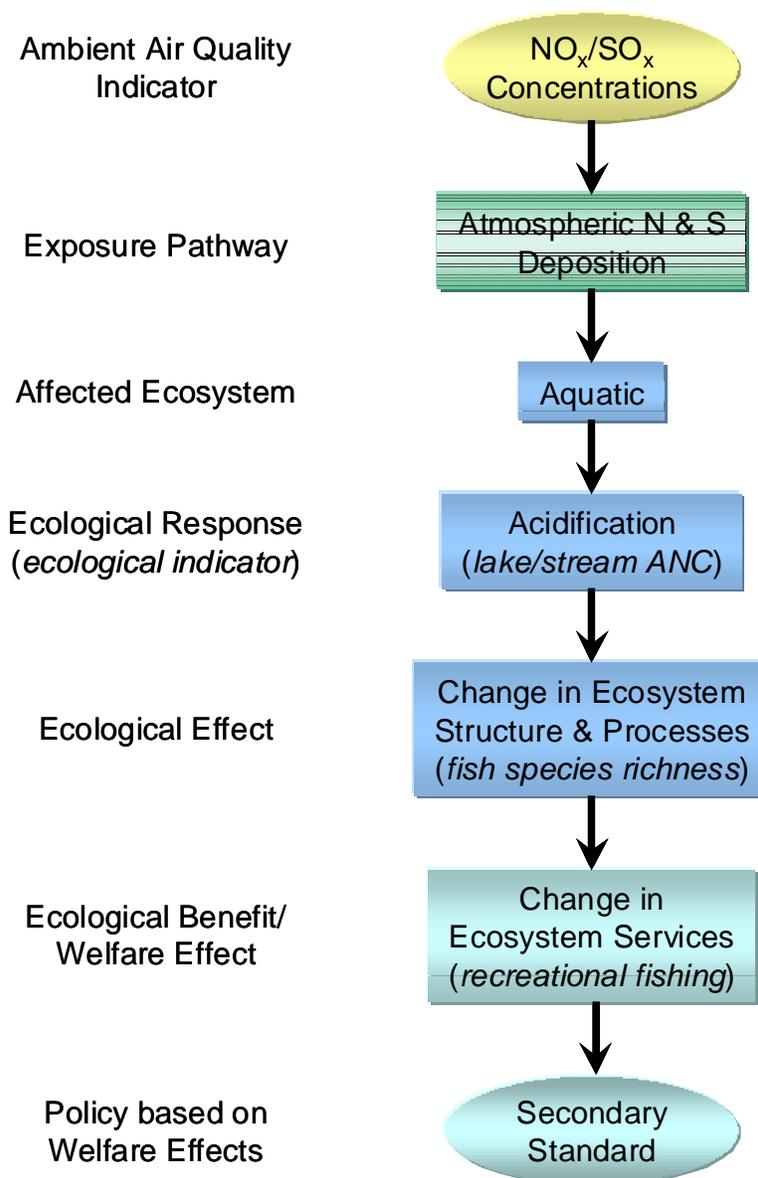
7 Valuing ecological benefits, or the contributions to social welfare derived from
 8 ecosystems, can be challenging, as noted in EPA’s *Ecological Benefits Assessment Strategic*
 9 *Plan* (U.S. EPA, 2006). It is necessary to recognize that in the analysis of the environmental
 10 responses associated with any particular policy or environmental management action, some of
 11 the ecosystem services likely to be affected are readily identified, whereas others will remain
 12 unidentified. Of those ecosystem services that are identified, some changes can be quantified,
 13 whereas others cannot. Within those services whose changes are quantified, only a few will
 14 likely be monetized, and many will remain unmonetized. Similar to health effects, only a portion
 15 of the ecosystem services affected by a policy can be monetized. The stepwise concept leading
 16 up to the valuation of ecosystems services is graphically depicted in **Figure 7-1**.



17
 18 **Figure 7-1.** Representation of the benefits assessment process indicating where
 19 some ecological benefits may remain unrecognized, unquantified, or unmonetized.
 20 (Source: U.S. EPA, 2006).

1 A conceptual model integrating the role of ecosystem services in characterizing known or
2 anticipated adverse effects to public welfare is shown in **Figure 7-2**. Under Section 108 of the
3 CAA, the secondary standard is to specify an acceptable level of the criteria pollutant(s) in the
4 ambient air that is protective of public welfare. For this review, the relevant air quality indicator
5 is interpreted as ambient NO_x and SO_x concentrations that can be linked to levels of deposition
6 for which there are adverse ecological effects. The air quality analyses described in Chapter 3
7 explore the sources, emissions, and deposition of total reactive nitrogen and sulfur and their
8 current contributions to ambient conditions. The case study analyses (described in Chapters 4
9 and 5) link deposition in sensitive ecosystems (e.g., the exposure pathway) to changes in a given
10 ecological indicator (e.g., for aquatic acidification, changes in acid neutralizing capacity [ANC])
11 and then to changes in ecosystems and the services they provide (e.g., fish species richness and
12 its influence on recreational fishing). To the extent possible for each targeted effect area, ambient
13 concentrations of nitrogen and sulfur (i.e., ambient air quality indicators) were linked to
14 deposition in sensitive ecosystems (i.e., exposure pathways), and then deposition was linked to
15 system response as measured by a given ecological indicator (e.g., lake and stream acidification
16 as measured by ANC). The ecological effect (e.g., changes in fish species richness, etc.) was
17 then, where possible, associated with changes in ecosystem services and their ecological benefits
18 or welfare effects (e.g., recreational fishing).

19 Knowledge about the relationships linking ambient concentrations and ecosystem
20 services can be used to inform a policy judgment on a known or anticipated adverse public
21 welfare effect. The conceptual model outlined for aquatic acidification in **Figure 7-2** can be
22 modified for any targeted effect area where sufficient data and models are available. For
23 example, a change in an ecosystem structure and process, such as foliar injury, would be
24 classified as an ecological effect, with the associated changes in ecosystem services, such as
25 primary productivity, food availability, and aesthetics (e.g., scenic viewing), classified as
26 ecological benefits/welfare effects. Alternatively, changes in biodiversity would be classified as
27 an ecological effect, and the associated changes in ecosystem services—productivity,
28 recreational viewing and aesthetics—would be classified as ecological benefits/welfare effects.
29 This information can then be used to characterize known or anticipated adverse effects to public
30 welfare and inform a policy based on welfare effects.



1
2 **Figure 7-2.** Conceptual model showing the relationships among ambient air
3 quality indicators and exposure pathways and the resulting impacts on
4 ecosystems, ecological responses, effects, and benefits to characterize known or
5 anticipated adverse effects to public welfare.

6 This chapter provides a concise summary of the information and evidence from the
7 *Integrated Science Assessment (ISA) for Oxides of Nitrogen and Sulfur–Ecological Criteria*
8 *(Final Report)* (ISA) (U.S. EPA, 2008) and the Risk and Exposure Assessment to characterize
9 known or anticipated adverse ecological effects from a scientific perspective.

1 **7.1 SUMMARY OF ECOLOGICAL RESPONSES, ECOLOGICAL**
2 **EFFECTS, AND WELFARE EFFECTS**

3 Summarizing the ecological responses (i.e., acidification and nutrient enrichment)
4 highlighted in the ISA (U.S. EPA, 2008, Section 3.2 and 3.3), the ecological effects (i.e., changes
5 in ecosystem structure and processes) explored by the Risk and Exposure Assessment, and the
6 ecological benefits/welfare effects described in this Risk and Exposure Assessment may clarify
7 which data are best suited to informing a policy based on welfare effects.

8 **7.1.1 Ecological Responses**

9 Deposition of nitrogen- and sulfur-containing compounds that are derived from NO_x and
10 SO_x may be wet (i.e., rain, snow), occult (i.e., cloud, fog), and dry (i.e., gases, particles) and can
11 affect ecosystem biogeochemistry and its community structure and function. Nitrogen and sulfur
12 interactions in the environment are highly complex. Both are essential and often limit the growth
13 or productivity and reproduction of ecosystems. Excess nitrogen (both oxidized and reduced
14 forms) or sulfur can lead to acidification and nutrient enrichment. Acidification causes a cascade
15 of effects that alter both terrestrial and aquatic ecosystems and that include slower growth, the
16 injury or death of forest vegetation and localized extinction of fish and other aquatic species.

17 With respect to acidification, the ISA (U.S. EPA, 2008, Section 3.2) determined

18 *The evidence is sufficient to infer a causal relationship between acidifying deposition and*
19 *effects on:*

20 *(1) biogeochemistry related to terrestrial and aquatic ecosystems;*

21 *(2) biota in terrestrial and aquatic ecosystems.*

22 In addition to acidification, NO_x acts with other forms of reactive nitrogen (including
23 reduced nitrogen) to increase the total amount of available nitrogen in ecosystems. Some of these
24 other forms of reactive nitrogen are part of atmospheric deposition; some are received via other
25 pathways. The contribution of nitrogen deposition to total nitrogen load varies among
26 ecosystems. Atmospheric nitrogen deposition is the main source of new nitrogen to most
27 headwater streams, high-elevation lakes, and low-order streams. Atmospheric nitrogen
28 deposition contributes varying proportions to the total nitrogen load in terrestrial, wetland,
29 freshwater, and estuarine ecosystems that receive nitrogen through multiple pathways (i.e.,
30 biological nitrogen-fixation, agricultural land runoff, and waste water effluent) (U.S. EPA, 2008,

1 Section 3.3). Nitrogen deposition alters numerous biogeochemical indicators, including primary
2 productivity that leads to changes in community composition and eutrophication.

3 With respect to nitrogen nutrient enrichment, the ISA (U.S. EPA, 2008, Section 3.3)
4 determined

5 *The evidence is sufficient to infer a causal relationship between nitrogen deposition, to*
6 *which NO_x and NH_x contribute, and the alteration of the following:*

7 *(1) biogeochemical cycling of nitrogen and carbon in terrestrial, wetland, freshwater*
8 *aquatic, and coastal marine ecosystems;*

9 *(2) biogenic flux of methane, and nitrous oxide in terrestrial and wetland ecosystems;*

10 *(3) species richness, species composition, and biodiversity in terrestrial, wetland,*
11 *freshwater aquatic and coastal marine ecosystems.*

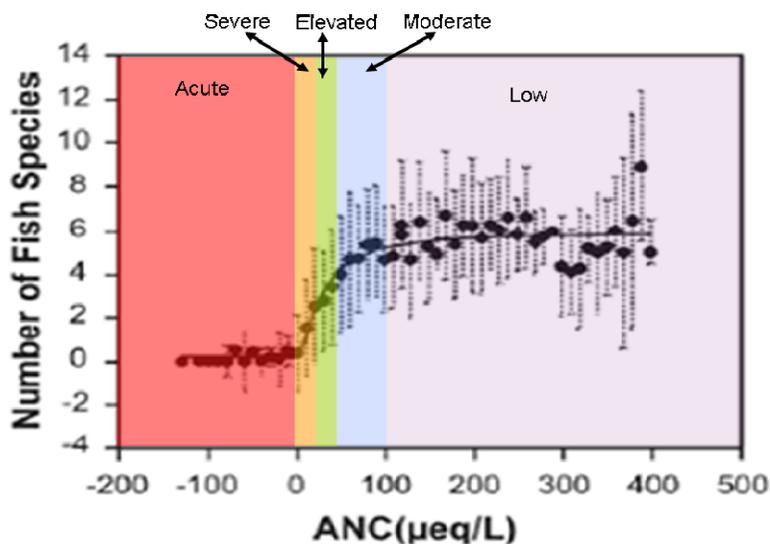
12 **7.1.2 Ecological Effects**

13 ***Aquatic Acidification***

14 The changes in ecosystem structure and processes associated with aquatic acidification
15 include changes in fish species richness as measured by the ecological indicator, ANC. The
16 impact of acidifying deposition on aquatic systems is controlled by several environmental
17 factors, such as geology, surface water flow, soil depth, and weathering rates, all of which
18 influence the ability of a watershed to neutralize the additional acidifying deposition and prevent
19 the lowering of surface ANC. ANC is a useful ecological indicator because it integrates the
20 overall acid-base status of a lake or stream and reflects how aquatic ecosystems respond to
21 acidifying deposition over time. There is also a relationship between ANC and the surface water
22 constituents that directly contribute to or ameliorate acidity-related stress, in particular,
23 concentrations of hydrogen ion (as pH), calcium (Ca²⁺), and aluminum (Al). In aquatic systems,
24 there is a direct relationship between ANC and fish and phyto-zooplankton diversity and
25 abundance (Baker et al., 1990).

26 The ANC of surface waters is widely used as a chemical indicator of acidic conditions
27 because it has been found in many studies to be the best single indicator of the biological
28 response and health of aquatic communities in acid-sensitive systems (Lien et al., 1992; Sullivan
29 et al., 2006). Logistic regression of species presence/absence data against ANC provides a
30 quantitative dose-response function, which indicates the probability of occurrence of an

1 organism for a given value of ANC. For example, the number of fish species present in a
 2 waterbody has been shown to be positively correlated with the ANC level in the water, with
 3 higher values supporting a greater richness and diversity of fish species (**Figure 7.1-1**). The
 4 diversity and distribution of phyto-zooplankton communities are also positively correlated with
 5 ANC.



6
 7 **Figure 7.1-1.** Number of fish species per lake or stream versus ANC level and
 8 aquatic status category (colored regions) for lakes in the Adirondack Case Study Area
 9 (Sullivan et al., 2006). The five aquatic status categories are described in **Table 7.1-1**.

10 For freshwater systems, ANC levels can be grouped into five major classes: <0, 0–20,
 11 20–50, 50–100, and >100 microequivalent per liter ($\mu\text{eq/L}$), with each range representing a
 12 probability of ecological damage to the community. ANC values >100 $\mu\text{eq/L}$ are generally not
 13 harmful (**Figure 7.1-1**) to biota. With ANC <100 $\mu\text{eq/L}$, fish fitness and community diversity
 14 begin to decline, but the overall health of the community remains high as long as ANC
 15 concentrations do not fall below 50 $\mu\text{eq/L}$. ANC concentrations <50 $\mu\text{eq/L}$ result in negative
 16 effects on sensitive biota. From 50 to 20 $\mu\text{eq/L}$, fish diversity and the overall fitness (i.e., health
 17 and reproduction) of most aquatic organisms in the waterbody are reduced. For ANC <20 $\mu\text{eq/L}$,
 18 all biota exhibit some level of negative effects, particularly because surface waters at this level
 19 are susceptible to episodic acidification and their associated harmful effects. Fish and plankton
 20 diversity and the structure of the communities continue to decline sharply to levels where
 21 acidophilic species begin to outnumber all other species. Below an ANC level of 0 $\mu\text{eq/L}$,
 22 complete loss of fish populations and extremely low diversity of planktonic communities occur.

1 At these low levels, only acidophilic species are present, but even their population and
 2 community structure are sharply reduced. The five categories of ANC and expected ecological
 3 effects are described in **Table 7.1-1** and are supported by a large body of research completed
 4 throughout the eastern United States (Sullivan et al., 2006).

5 **Table 7.1-1.** Aquatic Status Categories

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 forms. 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. 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.

6
 7 The maximum depositional load is the maximum amount of nitrogen and/or sulfur
 8 deposition that a given ecosystem can receive without the degradation of the ecological indicator
 9 for a targeted effect. In this case, the maximum deposition load is the acidic input of sulfur and
 10 nitrogen deposition that the watershed can neutralize and still maintain a given level of ANC in
 11 surface water. Based on the modeling described in Chapter 4 and Appendix 4, approximately
 12 50% of the 169 lakes modeled in the Adirondack Case Study Area are sensitive or at risk to
 13 acidifying deposition. For the 2002 model year, maximum depositional loads for ANC values of
 14 0, 20, 50, and 100 µeq/L were calculated. The exceedance value of a maximum depositional load

1 indicates the combined sulfur and nitrogen deposition in year 2002 that is greater than the
 2 amount of deposition the lake could buffer and still maintain the ANC level of above each of the
 3 four different ANC limits of 0, 20, 50, and 100 µeq/L. These data were extrapolated for the
 4 regional population of 1,849 lakes in the Adirondack Case Study Area that are from 0.5 to 2000
 5 hectares (ha) in size and at least 1 meter in depth, based on the Environmental Monitoring and
 6 Assessment Program (EMAP) Lake Probability Survey of 1991–1994. A similar analysis showed
 7 that approximately 75% of the 60 streams modeled in the Shenandoah Case Study Area are
 8 sensitive or at risk to acidifying deposition. For the year 2002, 52%, 72%, 85%, and 92% of the
 9 60 streams modeled received levels of combined total sulfur and total nitrogen deposition that
 10 exceeded maximum depositional loads of 0, 20, 50, and 100 µeq/L, respectively. The stream data
 11 for the Shenandoah Case Study Area cannot be extrapolated to a larger data set.

12 All of these data are shown in **Table 7.1-2**.

13 **Table 7.1-2.** Number and Percentage of Lakes in the Adirondack Case Study Area or Streams in
 14 the Shenandoah Case Study Area Currently Exceeding Maximum Depositional Loads Required
 15 to Maintain a Given ANC.

	ANC Limit 100 µeq/L		ANC Limit 50 µeq/L		ANC Limit 20 µeq/L		ANC Limit 0 µeq/L	
	No. Lakes	% Lakes	No. Lakes	% Lakes	No. Lakes	% Lakes	No. Lakes	% Lakes
Adirondack Case Study Area Lakes								
N=169 (Modeled for case study)	98	58	74	44	47	28	30	18
N=1,849 (Regional population)	951	51	666	36	242	13	135	7
Shenandoah Case Study Area Streams	No. Streams	% Streams	No. Streams	% Streams	No. Streams	% Streams	No. Streams	% Streams
N=60 (Modeled for case study)	55	92	51	85	43	72	31	52

16
 17 During the year, ANC values can fluctuate and are driven by pulses of acidity due to
 18 atmospheric deposition. Episodic acidification events occur during spring snowmelt when
 19 accumulated deposition is flushed into surface waters. In general, summer ANC values are

1 approximately 30 $\mu\text{eq/L}$ higher in summer than in fall ANC values (Stoddard et al., 2003).
2 Detrimental ecological effects can occur when ANC levels drop into the 20 $\mu\text{eq/L}$ range where
3 reproductive effects occur. If ANC drops $<0 \mu\text{eq/L}$, it is deadly to fish populations. The higher
4 the ANC level, the more ecological protection there is against episodic events. For example,
5 exposure to an annual mean ANC of 50 $\mu\text{eq/L}$ can be protective against low pulses of acidity.
6 The length of time a fish population can survive an episodic acid pulse varies depending on the
7 habitat, stream conditions, and other ecological factors.

8 ***Terrestrial Acidification***

9 The ecological effects associated with terrestrial acidification include increasing
10 concentrations of nitrogen and sulfur in the soil, which accelerate the leaching of sulfate (SO_4^{2-})
11 and nitrate (NO_3^-) from the soil to drainage water. Atmospheric deposition of sulfur and nitrogen
12 provides anions that are more mobile in the soil environment than are naturally occurring anions
13 in the soil; these mineral acid anions can accelerate natural rates of base-cation leaching,
14 particularly Ca^{2+} and magnesium (Mg^{2+}), leading to increased mobilization of inorganic Al,
15 which is toxic to tree roots. Acidifying deposition can also affect terrestrial ecosystems by
16 causing direct phytotoxic effects on plant foliage.

17 The tree species most sensitive to soil acidification due to atmospheric nitrogen and
18 sulfur deposition include red spruce (i.e., *Picea rubens*, a coniferous tree species) and sugar
19 maple (i.e., *Acer saccharum*, a deciduous tree species). Much of the scientific literature
20 discussing terrestrial soil acidification focuses on Ca^{2+} depletion and Al mobilization as the
21 primary indicators of detrimental effects on terrestrial vegetation. Both of these indicators are
22 strongly influenced by soil acidification, and both have been shown to have quantitative links to
23 vegetation growth and vigor.

24 This review focused on sugar maple and red spruce because they occur in areas that
25 receive high acidifying deposition and are known to be negatively affected by Ca^{2+} depletion
26 and high concentrations of Al, as measured by base cation to aluminum (Bc/Al) ratios in soils.
27 The ecological effects associated with acidifying deposition are summarized in **Table 7.1-3**.
28 Sugar maple and red spruce abundance and growth (i.e., crown vigor, biomass) were linked
29 quantitatively to acidification symptoms. Bc/Al ratios in soils were selected as the indicator to

1 evaluate acidifying deposition loadings in terrestrial systems using the U.S. Forest Service Forest
 2 Inventory and Analysis (FIA) database.

3 **Table 7.1-3.** Summary of Linkages among Acidifying Deposition, Biogeochemical Processes
 4 that Affect Ca^{2+} , Physiological Processes That Are Influenced by Ca^{2+} , and Effect on Forest
 5 Function

Biogeochemical Response to Acidifying Deposition	Physiological Response	Effect on Forest Function
Leach Ca^{2+} from leaf membrane	Reduce the cold tolerance of needles in red spruce	Loss of current year needles in red spruce
Reduce the ratio of Ca^{2+}/Al in soil and soil solutions	Dysfunction in fine roots of red spruce blocks uptake of Ca^{2+}	Decreased growth and increased susceptibility to stress in red spruce
Reduce the ratio of Ca^{2+}/Al in soil and soil solutions	More energy is used to acquire Ca^{2+} in soils with low Ca^{2+}/Al ratios	Decreased growth and increased photosynthetic allocation to red spruce roots
Reduce the availability of nutrient cations in marginal soils	Sugar maples on drought-prone or nutrient-poor soils are less able to withstand stresses	Episodic dieback and growth impairment in sugar maple

6 **Source:** Fenn et al., 2006.

7 Three values of the indicator were used to calculate critical loads of $(\text{Bc}/\text{Al})_{\text{crit}}$, which
 8 represent different levels of tree protection associated with total nitrogen and sulfur deposition:
 9 0.6, 1.2, and 10 (Section 4.3.1.2). The $(\text{Bc}/\text{Al})_{\text{crit}}$ ratio of 0.6 represents the highest level of
 10 impact (i.e., lowest level of protection) to tree health and growth and was selected because 75%
 11 of species found growing in North America experience reduced growth at this Bc/Al ratio
 12 (Sverdrup and Warfvinge, 1993; see Figure 4.3-1). In addition, a soil solution Bc/Al ratio of 0.6
 13 has been linked to a 20% and 35% reduction in sugar maple and red spruce growth, respectively
 14 (Sverdrup and Warfringe, 1993). The $(\text{Bc}/\text{Al})_{\text{crit}}$ ratio of 1.2 is considered to represent a moderate
 15 level of impact, as the growth of 50% of tree species (found growing in North America) were
 16 negatively impacted at this soil solution ratio. The $(\text{Bc}/\text{Al})_{\text{crit}}$ ratio of 10.0 represents the lowest
 17 level of impact (i.e., greatest level of protection) to tree growth; it is the most conservative value
 18 used in studies in the United States and Canada (NEG/ECP, 2001; McNulty et al., 2007;
 19 Watmough et al., 2004).

20 Critical loads for 2002 were calculated for multiple areas in 24 states for sugar maple and
 21 in 8 states for red spruce based on the FIA database. The exceedance value of a critical load
 22 indicates the combined total sulfur and total nitrogen deposition in year 2002 that is greater than

1 the amount of deposition forest soils could buffer and still maintain the Bc/A1 level of above
 2 each of the three different Bc/A1 limits of 0.6, 1.2, and 10. These data are summarized in
 3 **Table 7.1-4.**

4 **Table 7.1-4.** Percent of forest plots in the range of sugar maple and res spruce currently
 5 exceeding critical loads required to maintain a given Bc/A1.

	Bc/A1 = 0.6	Bc/A1 = 1.0	Bc/A1 = 10
Sugar Maple (n=4,992; 24 states)	3	12	75
Red spruce (n=763; 8 states)	3	5	36

6

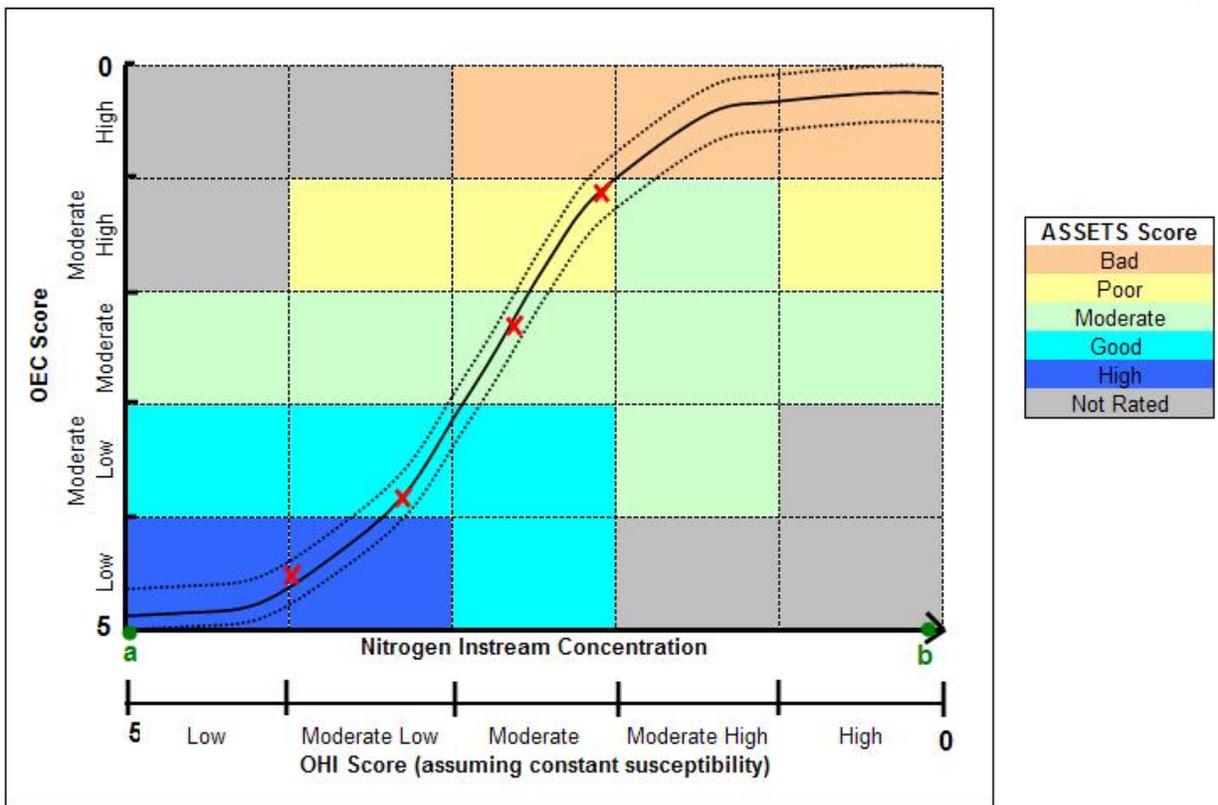
7 ***Aquatic Nutrient Enrichment***

8 Nitrogen is an essential nutrient for aquatic ecosystem fertility, including lake, marine,
 9 and estuarine ecosystems, and is often the limiting nutrient for growth and reproduction in many
 10 of these ecosystems. Nutrient enrichment may have beneficial fertilization effects, but can also
 11 lead to over-enrichment of a system, causing eutrophication. Excessive nutrient enrichment can
 12 result in changes to ecosystem structure and function by causing harmful algal blooms, hypoxia
 13 (i.e., reduced dissolved oxygen), anoxia (i.e., absence of dissolved oxygen), fish kills, habitat
 14 degradation, and decreases in biodiversity. In addition, aquatic ecosystems are impacted by both
 15 oxidized and reduced forms of nitrogen, not oxidized nitrogen alone, which is the regulated
 16 criteria pollutant.

17 Because of the cascading impacts and effects of nutrient enrichment, there are a suite of
 18 possible ecological indicators. The National Oceanic and Atmospheric Administration’s
 19 (NOAA) National Estuarine Eutrophication Assessment (NEEA) Update provides a detailed
 20 explanation of the biological indicators used to evaluate eutrophic status. Five biological
 21 indicators are used in this index: chlorophyll *a*, macroalgae, dissolved oxygen, nuisance/toxic
 22 algal blooms, and submerged aquatic vegetation. NOAA’s Assessment of Estuarine Trophic
 23 Status (ASSETS) produces an eutrophication index (EI) that is an estimation of the likelihood
 24 that the estuary is experiencing eutrophication or will experience eutrophication in the future
 25 based on the five indicators listed above.

26 In this assessment, two main stem rivers, the Potomac River and the Neuse River, were
 27 selected to analyze the influence of total atmospheric nitrogen deposition on eutrophic conditions
 28 to the Chesapeake Bay and Pamlico Sound, respectively. The EI for both of these estuaries is

1 currently “Bad.” Response curves were developed (see **Figure 7.1-2**) that related instream
 2 nitrogen levels to the EI. The curves were used to determine the instream nitrogen concentrations
 3 necessary to move the EI from a score of “Bad” to “Poor,” a one-category improvement. The
 4 instream nitrogen concentrations can be used to back-calculate the required decrease in
 5 atmospheric deposition to achieve those concentrations. For both estuaries, a 100% or greater
 6 reduction in atmospheric deposition was necessary, demonstrating that reductions in additional
 7 sources of nitrogen loading to the estuaries are also required. See Chapter 5 and Appendix 6 for
 8 additional details on the methods and results of the Potomac River/Potomac Estuary Case Study
 9 and the Neuse River/Neuse River Estuary Case Study.



10
 11 **Figure 7.1-2.** ASSETS EI response curve. Point “a” represents the background
 12 nitrogen concentration that would occur in the system with no anthropogenic inputs
 13 (assuming the system is not naturally eutrophic) or the system at a pristine state. The
 14 upper bound of the instream total nitrogen concentration, Point “b,” is the maximum
 15 nitrogen concentration at which the system is nitrogen-limited; above this point, the
 16 nitrogen inputs to the system no longer affect the eutrophication condition.

17 In addition to the case studies for the Potomac and Neuse River estuaries, the ISA (U.S.
 18 EPA, 2008) presents scientific studies that show increased atmospheric nitrogen deposition in

1 high alpine lakes and streams can cause a shift in community composition and reduce algal
2 biodiversity. Elevated nitrogen deposition results in changes in algal species composition,
3 especially in sensitive oligotrophic lakes. Two opportunistic diatom species, *Asterionella*
4 *formosa* and *Fragilaria crotonensis* (McKnight et al., 1990; Lafrancois et al., 2004; Saros, 2005)
5 now dominate the flora of at least several alpine and montane Rocky Mountain lakes, with
6 similar field data showing shifts in dominant algal species in other parts of the western United
7 States. A hindcasting exercise has concluded that the change occurred in Rocky Mountain
8 National Park lake algae between 1850 and 1964 was associated with an increase of only about
9 1.5 kg N/ha in wet nitrogen deposition. (Baron, 2006). Similar changes inferred from lake
10 sediment cores of the Beartooth Mountains of Wyoming also occurred in about 1.5 kg N/ha
11 deposition (Saros et al., 2003). A strong relationship exists between aquatic eutrophication of
12 high alpine lakes in the Rocky Mountains and atmospheric deposition because atmospheric
13 deposition is the only source of nitrogen to these systems.

14 ***Terrestrial Nutrient Enrichment***

15 Terrestrial ecosystems typically respond to both oxidized and reduced forms of nitrogen,
16 not oxidized nitrogen alone, which is the regulated criteria pollutant. Excess nitrogen in
17 terrestrial ecosystems changes ecosystem structure and processes by inadvertent fertilization of
18 vegetation, creating nutrient imbalances and increased growth rates in some species over others,
19 which changes competitive interactions among species. These impacts ultimately reduce
20 ecosystem health and biodiversity. For example, forest growth enhancement can potentially
21 exacerbate other nutrient deficiencies, such as calcium (Ca^{2+}), Mg^{2+} , or potassium (K^{+}), thereby
22 causing compared forest health declines. Enhanced growth generally occurs above ground level
23 (stems and leaves), producing more shoot-growth compared to root growth. this increase in the
24 “shoot to root” ratio can cause decreased resistance to environmental stressors, such as drought
25 (Grulke et al., 1998). In conifer species, multiple long-term experiments (see Appendix 7) have
26 demonstrated transient growth increases (generally at deposition rates <10 kg N/ha/yr) followed
27 by increased mortality, especially at higher rates of fertilization (Grukke et al., 1998; Takemoto et
28 al., 2001). In western conifer forests, atmospheric nitrogen deposition has been shown to cause
29 increased litter accumulation on the ground surface and carbon storage in aboveground portion
30 of woody biomass of these, which, in turn, may lead to increased susceptibility to more severe

1 fires (Grukle et al., 2008). Grassland communities that are adapted to low nutrient supply can
2 exhibit substantial sensitivity to nutrient enrichment effects of nitrogen deposition. Invasive
3 species of grass that may have been suppressed by nitrogen limitation can now better compete
4 and alter species dominance.

5 Two of the primary indicators of nitrogen over-enrichment in forested watersheds are the
6 leaching of NO_3^- in soil drainage waters and the export of NO_3^- in stream water (Fenn et al.,
7 2008). Low carbon to nitrogen (C:N) ratios in soils are also commonly related to increased
8 nitrification, potential increases in soil acidity, and releases of NO_3^- to receiving waters;
9 however, these measurements are not always widely available.

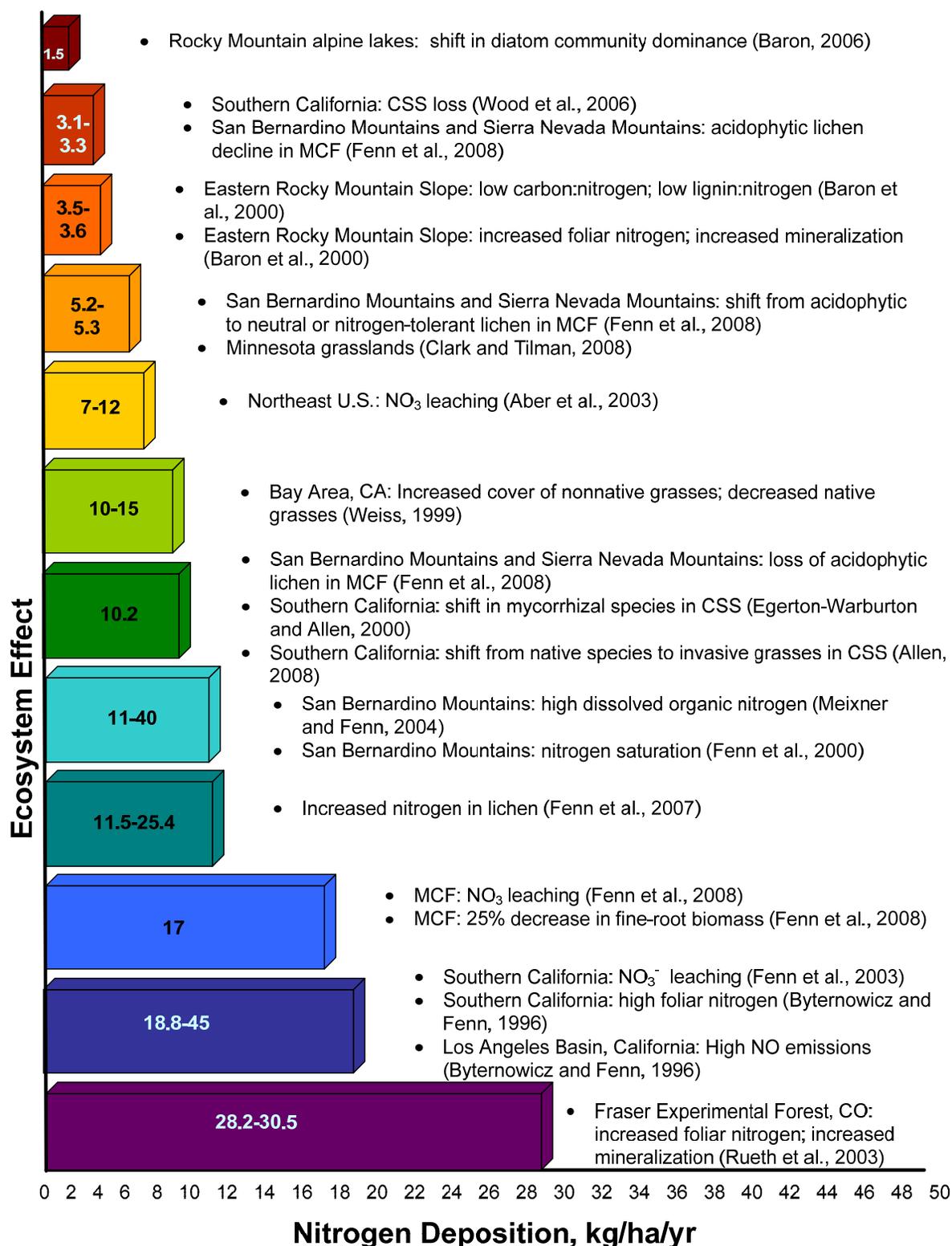
10 Because of data limitations, the assessment of ecological effects was based on a
11 qualitative weight of evidence approach that used the current scientific literature to determine
12 benchmark values for ecological effects attributable to nitrogen deposition in southern California
13 coastal sage scrub (CSS) and mixed conifer forest (MCF) communities in the Sierra Nevada and
14 San Bernardino mountains of California(see **7.1-3**). There are sufficient data to relate an
15 ecological effect to atmospheric nitrogen deposition. For the CSS community, the following
16 ecological thresholds were identified:

- 17 ▪ 3.3 kg N/ha/yr — the amount of nitrogen uptake by a vigorous stand of CSS; above this
18 level, nitrogen may no longer be limiting
- 19 ▪ 10 kg N/ha/yr — mycorrhizal community changes, CSS decline.

20 For the MCF community in the Pacific Coast states, the following ecological thresholds
21 were identified:

- 22 ▪ 3.1 kg N/ha/yr — shift from sensitive to tolerant lichen species
- 23 ▪ 5.2 kg N/ha/yr — dominance of the tolerant lichen species
- 24 ▪ 10.2 kg N/ha/yr — loss of sensitive lichen species
- 25 ▪ 17 kg N/ha/yr — leaching of NO_3^- into streams.

26 The deposition loads used are presented in **Figure 7.1-3**.



1
2 **Figure 7.1-3.** Total Atmospheric nitrogen deposition loads for several ecological effects,
3 including California Coastal Sage Scrub, Pacific coast Mixed Conifer Forest, and Pacific
4 Northwest lichen.

1 **7.1.3 Ecological Benefits/Welfare Effects**

2 As shown in **Figure 7-1**, some of the ecological benefits or welfare effects as described
3 by changes in ecosystem services are readily identified, whereas others are not. Of those
4 ecosystem services that are identified, some changes can be quantified, whereas others can not.
5 Within those services whose changes are quantified, only a few can be monetized, and many will
6 remain unmonetized. A brief description of the ecosystem services identified for each ecological
7 response is provided below, including quantification and monetization, where possible.

8 ***Aquatic Acidification***

9 The connection between changes in ecological effects associated with declining ANC
10 levels (see **Figure 7.1-1** and **Table 7.1-1**) and changes in ecosystem services may aid in
11 determining adverse impacts to public welfare. Examples of these ecosystem services include
12 recreational and subsistence fishing and natural habitat provisioning.

13 Food and fresh water are generally the most important provisioning services provided by
14 inland surface waters (MEA, 2005). There are relatively few data available for measuring the
15 effects of acidification on subsistence and other consumers.

16 The cultural service that is likely to be most widely and significantly affected by aquatic
17 acidification is recreational fishing because it depends directly on the health and abundance of
18 aquatic wildlife. For the 17 states included in the northeastern region of the United States, by the
19 Department of the Interior, and roughly corresponding to the sugar maple and red spruce range,
20 in the 2006 National Survey of Fishing, Hunting, and Wildlife Associated Recreation (FHWAR):

- 21 ▪ More than 9% of adults in this part of the country participate annually in freshwater
22 (excluding Great Lakes) fishing.
- 23 ▪ The total number of freshwater fishing days was 140.8 million.
- 24 ▪ Roughly two-thirds of these fishing days were at ponds, lakes, or reservoirs in these states.
- 25 ▪ There were estimated average consumer surplus values per day of \$35.91 for recreational
26 fishing (Kaval and Loomis, 2003).
- 27 ▪ The implied total annual value was \$5 billion.

28 Freshwater ecosystems also provide biological control services (regulating services) by
29 providing environments that sustain delicate aquatic food chains. Some of these services may be
30 captured through measures of provisioning and cultural services. For example, these biological

1 control services may serve as “intermediate” inputs that support the production of “final”
2 recreational fishing and other cultural services.

3 ***Terrestrial Acidification***

4 The ecosystem services affected by NO_x and SO_x deposition and examined in this Risk
5 and Exposure Assessment include provisioning services (i.e., timber and syrup production),
6 cultural services (i.e., recreation, aesthetic, tourism) and regulating services (i.e., soil
7 stabilization and erosion control, water regulation, climate regulation).

8 Provisioning services are as follows:

- 9 ■ Saw timber—1.2 billion board feet (combined sugar maple and red spruce)
- 10 ■ Syrup—1.2 million and 1.4 million gallons; 19% of worldwide production.

11 Cultural services are as follows:

- 12 ■ Thirty-one percent (31%) of the United States adult population (16 and older) visited a
13 wilderness or primitive area during the previous year.
- 14 ■ Thirty-two percent (32%) engaged in day hiking (Cordell et al., n.d.).
- 15 ■ Sixteen percent (16%) of adults in the northeastern United States¹ participated in off-road
16 vehicle recreation, for an implied value of \$9.25 billion.
- 17 ■ Five and one-half percent (5.5%) of adults in the northeastern United States participated in
18 hunting; the total number of hunting days occurring in those states was 83.8 million, for an
19 implied value of \$4.38 billion.
- 20 ■ Ten percent (10%) of adults in the northeastern United States participated in wildlife
21 viewing away from home, for an implied value of \$4.21 billion.
- 22 ■ Thirty percent (30%) of residents (Great Lakes area) engaged in tourism for fall color
23 viewing.
- 24 ■ Twenty-two percent (22%) of households visiting Vermont in 2001 made the trip primarily
25 for the purpose of viewing fall colors.

26 Regulating services are as follows:

- 27 ■ Regulation of soil erosion, runoff, and sedimentation that can adversely impact surface
28 waters
- 29 ■ Storage and regulation of the *quantity* and flows of water in watersheds
- 30 ■ Regulation of climate locally by trapping moisture and globally by sequestering carbon.

¹ This area includes Connecticut, Delaware, District of Columbia, Illinois, Indiana, Maine, Maryland, Massachusetts, Michigan, New Hampshire, New Jersey, New York, Ohio, Pennsylvania, Rhode Island, Vermont, West Virginia, and Wisconsin.

1 The changes to these services as a result of acidification are difficult to quantify;
2 however, an analysis of the change in sugar maple timber production has been completed and the
3 results can be found in Appendix 8.

4 ***Aquatic Nutrient Enrichment***

5 While the ASSETS EI does not provide a direct link to ecological benefits provided by an
6 ecosystem, the impacts of the indicators contributing to the ASSETS EI may be directly related
7 to ecosystem services. For instance, if the indicator is dissolved oxygen, the ecological endpoint
8 or impact of having low dissolved oxygen is a decrease in the populations of fish that are highly
9 sensitive to dissolved oxygen conditions. Decreases in fish populations can be evaluated by the
10 ecosystem services they impact, namely commercial and recreational fishing.

11 In addition, nutrient enrichment affects other recreational services, including beach use,
12 boating, bird watching, and non-use (also known as existence) services. These activities account
13 for a total of nearly 300 million activity days and at least \$3.3 billion (in 2007 dollars) of
14 consumer surplus value in the Chesapeake Bay area. The regulating services supported by
15 estuaries and marshes (i.e., climate, biological, erosion prevention, and protection against natural
16 hazards) have the potential for very large economic value; however, it is difficult to identify and
17 quantify the effect of nutrient loadings on these services.

18 ***Terrestrial Nutrient Enrichment***

19 The ecosystem service impacts of terrestrial nutrient enrichment include primarily
20 cultural and regulating services. In CSS, nutrient enrichment is associated with a decline in CSS
21 and an increase in nonnative grasses and other species, reduced viability of threatened and
22 endangered species associated with CSS, and an increase in fire frequency. In MCF, nutrient
23 enrichment is associated with changes in habitat suitability and increased tree mortality,
24 increased fire intensity, and a change in the forest's nutrient cycling that may affect surface water
25 quality through increased NO_3^- leaching (U.S. EPA, 2008).

26 The primary cultural ecosystem services associated with CSS and MCF are recreation,
27 aesthetic, and nonuse values. CSS and MCF are found in numerous recreation areas in
28 California. Seven national parks and monuments in California contain CSS or MCF, and these
29 ecosystems are considered part of the attraction. They are the iconic ecosystems of California
30 and are examples of the pre-development environment. Together, more than 6.7 million visitors

1 traveled through these parks in 2008. In addition, numerous state and county parks encompass
2 CSS and MCF ecosystems For example, California’s Torrey Pines State natural Reserve
3 specifically protects CSS habitat (see <http://www.torreypine.org/>). Visitors to these parks engage
4 in activities such as camping, hiking, attending educational programs, horseback riding, wildlife
5 viewing, water-based recreation, and fishing. CSS and MCF provide the environment for these
6 activities and habitat fro the wildlife that visitors come to view. Declines in these ecosystems
7 mean less wildlife and fewer wilderness areas in which to participate in outdoor activity.

8 The 2006 Fishing, Hunting, and Wildlife-Associated Recreation for California (DOI,
9 2007) reports on the number of individuals involved in fishing, hunting, and wildlife viewing in
10 California. According to this survey, the number of activity days for these forms of recreation
11 was more than 67 million days in 2007. The total benefits in 2006 from fishing, hunting, and
12 wildlife viewing away from home in California were approximately \$947 million, \$169 million,
13 and \$3.59 billion, respectively, based on average values from Kaval and Loomis (2003). Other
14 data indicate that the aggregate annual benefit for California residents from trail hiking in 2007
15 was \$11.59 billion.

16 Beyond the recreational value, the CSS landscape and the MCF provide aesthetic services
17 to local residents and homeowners who live near CSS or MCF. Nonuse value, also called
18 existence value or preservation value, encompasses a variety of motivations that lead individuals
19 to place value on environmental goods or services that they do not use. Communities of CSS are
20 home to three important federally endangered species. MCF is home to one federally endangered
21 species and a number of state-level sensitive species. The Audubon Society lists 28 important
22 bird areas in CSS habitats and at least 5 in MCF in California
23 (<http://ca.audubon.org/iba/index.shtml>).²

24 The Terrestrial Nutrient Enrichment Case Study identified fire regulation as a service that
25 encourages growth of more flammable grass species in nutrient enriched CSS and MCF
26 ecosystems. Over the 5-year period from 2004 to 2008, Southern California experienced, on
27 average, more than 4,000 fires a year burning, on average, more than 400,000 acres (National
28 Association of State Foresters [NASF], 2009). Improved fire regulation leads to short-term and
29 long-term benefits. The California Department of Forestry and Fire Protection (CAL FIRE)
30 estimated that the average annual cost for loss of homes due to wildfires was \$163 million per

² Important bird areas are sites that provide essential habitats for one or more species of birds.

1 year from 1984 to 1994 (CAL FIRE, 1996) and more than \$250 million in 2007 (CAL FIRE,
2 2008). In fiscal year 2008, CAL FIRE’s cost for fire suppression activities was nearly \$300
3 million (CAL FIRE, 2008). Therefore, even a 1% reduction in these damages and costs would
4 imply benefits of more than \$5 million per year. In the long term, decreased frequency of fires
5 could result in an increase in property values in fire-prone areas.

6 In the MCF Case Study Area, maintaining water quality emerged as a regulating service
7 that can be upset by excessive nitrogen. When the soil becomes saturated, nitrates may leach into
8 the surface water and cause acidification. Several large rivers and Lake Tahoe cut through MCF
9 areas. Additional nitrogen from MCF areas could further degrade waters that are already stressed
10 by numerous other sources of nutrients and pollution.

11 **7.2 UNCERTAINTY**

12 To aid in informing a policy decision regarding adverse effects to public welfare
13 associated with acidification and nitrogen nutrient enrichment, it is necessary considering the
14 variability and uncertainty associated with each step of the conceptual model describing the
15 relationships between ambient air concentrations and developing a policy based on the welfare
16 effects shown in **Figure 7-2**. The result of the cascade of relationships shown in **Figure 7-2** can
17 be used to assess the scientific basis for characterizing known or anticipated adverse public
18 welfare effects. It is recognized that relative uncertainties and variability exist within each box
19 along this continuum as information is passed along to the next step, and they are described in
20 the following paragraphs.

21 **Ambient Air Quality Indicator:** Monitoring networks that provide routine ambient
22 measurements of NO_x and SO_x have limited coverage for sensitive ecosystems. In view of the
23 lack of measured air concentrations, NO_x and SO_x concentration estimates were used from an air
24 quality photochemical dispersion model, the Community Multiscale Air Quality (CMAQ)
25 model. This model-based approach leads to some uncertainties in the estimates of NO_x and SO_x
26 in the case study areas and large assessment areas. The uncertainties lie in model formation,
27 model inputs, and spatial scale. Model formulation uncertainties most relevant for this
28 assessment include aspects of the nonlinear photochemical processes, which determine the
29 chemical form and transformations of NO_x component species and SO_x, in the atmosphere over
30 multiday time periods, and processes that affect the removal of NO_x and SO_x through deposition.

1 The NO_x and SO_x emissions inputs to the air quality model for some source categories contain
2 uncertainties in part because they are not “measured,” but are instead based on emissions models
3 that rely on category-specific emissions factors and a mix of national, regional, and local activity
4 data. However, emissions from electric generating units (EGUs) that emit large amounts of NO_x
5 and SO_x are based on measurements using continuous emissions monitoring devices. The key
6 uncertainties in meteorological inputs to the air quality model that affect NO_x and SO_x
7 concentrations may be associated with the ability of meteorological models to adequately capture
8 the degree of vertical mixing, the extent of clouds, and the amount of precipitation under certain
9 meteorological conditions and in complex terrain. Finally, for this analysis, CMAQ model
10 simulations with a horizontal resolution of approximately 12 × 12 km were used. While fairly
11 refined, this resolution may be too coarse to resolve the gradients in the concentrations of NO_x
12 and SO_x in and near all ecosystems.

13 **Exposure Pathway:** There are known uncertainties associated with the CMAQ-modeled
14 dry deposition estimates and the National Atmospheric Deposition Program (NADP)–measured
15 wet deposition estimates. These estimates are based on the best information available. CMAQ is
16 a state-of-the-science air quality model, and the monitored NADP data is very robust. Merging
17 the modeled and monitored data helps offset the uncertainty inherently present in each dataset.
18 Overall, there is confidence in the relative magnitude of these estimates.

19 **Affected Ecosystem:** Translating the deposition estimates to aquatic and terrestrial
20 ecosystems involves uncertainty related to environmental characteristics such as geology,
21 weathering rates, soil type, and other climatic conditions. This applies to both aquatic (e.g., lake
22 and watershed characteristics) and terrestrial (e.g., forest type, soil characteristics) ecosystems.

23 In selecting the case study areas, those areas highlighted in the ISA (U.S. EPA, 2008) for
24 which there was the best available data and models were relied upon. The areas were chosen
25 based on 1) known sensitivity using ecological characteristics and 2) vulnerability to effects
26 under current levels of NO_x and SO_x deposition. In selecting the areas, the results are most likely
27 not biased downward, but there is also uncertainty that the most sensitive ecosystems in the
28 contiguous United States were selected. Therefore, when extrapolating the case study results to
29 larger areas, it is important to recognize that these responses may not be applicable to other
30 ecosystems with differing characteristics. In scaling up to larger assessment areas, these
31 extrapolations were based on known habitat and responses rather than scaling up nationwide. For

1 example, the Adirondack lakes sampled in the TIME/LTM network were chosen for their
2 representativeness of a larger suite of lakes in the Adirondack Case Study Area.

3 **Ecological Response:** The ecological responses focused on in this review are
4 acidification and nutrient enrichment. These responses were measured by the ecological
5 indicators chosen for this Risk and Exposure Assessment. Based on the analyses presented in
6 Chapters 4 and 5 and in the case study reports (Appendices 4 through 7), there is confidence
7 about several relationships. In general, there is a strong relationship between aquatic acidification
8 and ANC. There is also a good relationship between terrestrial acidification and the Bc/Al ratio.
9 There is a weak relationship between aquatic nitrogen nutrient enrichment and the ASSETS EI in
10 terms of the atmospheric contribution because of the many other non-atmospheric contributions
11 to aquatic nutrient enrichment. However, a strong relationship exists between atmospheric
12 deposition of nitrogen and ecological responses and effects in high alpine lakes in the Rocky
13 Mountains because atmospheric deposition is the only source of nitrogen to these systems. There
14 is a positive, qualitative relationship between terrestrial nitrogen nutrient enrichment and
15 atmospheric nitrogen deposition in terms of shifts to nitrogen-tolerant species and, ultimately,
16 NO_3^- leaching from terrestrial to aquatic ecosystems.

17 **Ecological Effect:** Changes in ecosystem structure and processes are measures of
18 ecological effects and may be described as physical, chemical, and biological activities that
19 influence the flows, storage, and transformation of material and energy within and through
20 ecosystems (U.S. EPA, 2006). For aquatic acidification, there is a strong relationship between
21 fish species richness and ANC levels (see **Figure 7.1-1** and **Table 7.1-1**). For terrestrial
22 acidification, there is also a strong relationship between soil acidification and Bc/Al ratio (see
23 **Table 7.1-3**). The relationship between changes in ecosystem structure and process and
24 ecological effects is less certain for aquatic nitrogen nutrient enrichment because of
25 complications from other nonatmospheric sources of nitrogen and the contribution of both
26 oxidized and reduced nitrogen to enrichment effects. Both oxidized and reduced forms of
27 nitrogen also contribute to ecological effects due to terrestrial nitrogen nutrient enrichment as
28 measured by atmospheric deposition of total reactive nitrogen. This relationship is based on
29 qualitative, weight-of-evidence approaches and on the scientific literature highlighted in the ISA
30 (U.S. EPA, 2008) (see **Figure 7.1-3**)

1 **Ecological Benefit/Welfare Effects:** The next step in the conceptual model is to describe
2 a change in ecosystem services related to a given ecological benefit or welfare effect. According
3 to EPA’s *Ecological Benefits Assessment Strategic Plan* (U.S. EPA, 2006), ecological benefits
4 are contributions to social welfare of ecological goods and services, and, specifically to net
5 improvements in social welfare that result from changes in the quantity or quality of ecological
6 goods and services attributable to EPA policy. Overall, there is less confidence in the current
7 ability to translate the changes in ecological effects to changes in ecosystem services than in the
8 ability to quantify changes in ecosystem structure and processes.

9 However, within this context, there is a strong relationship between fish species richness
10 and recreational fishing and their association with aquatic acidification in the Adirondack Case
11 Study Area. Additional services that are impacted by changes in ecosystem structure and
12 processes that are associated with aquatic acidification are subsistence fishing, natural habitat
13 provision, and biological control services that sustain aquatic food chains. For terrestrial
14 acidification, a number of changes in ecosystem services were identified, including provisioning
15 services (e.g., timber and syrup production), cultural services (e.g., recreation, aesthetic, tourism)
16 and regulating services (e.g., soil stabilization and erosion control, water regulation, climate
17 regulation).

18 The relationships among the ecological benefits associated with the changes to ecosystem
19 structure and processes for aquatic nutrient enrichment are less certain. Qualitatively, nitrogen
20 nutrient enrichment impacts recreational services such as beach use, boating, and bird watching,
21 as well as regulating services. Terrestrial nitrogen nutrient enrichment is qualitatively associated
22 with a number of ecosystem services, including species shifts and changes in habitat. Additional
23 ecosystem services affected include recreation, aesthetics, and wildlife preservation, as well as
24 fire regulation.

25 **7.3 CONCLUSIONS**

26 Although many uncertainties are associated with these analyses, there is confidence from
27 a scientific perspective that known or anticipated adverse ecological effects are occurring under
28 current ambient loadings of nitrogen and sulfur in sensitive ecosystems across the United States.
29 Of all the case study analyses, the most confidence lies in the ecological responses, effects, and
30 benefits associated with aquatic acidification, and there is a fair amount of confidence about

1 those associated with terrestrial acidification. The least amount of confidence is in the ecological
2 responses, effects, and benefits associated with aquatic nitrogen nutrient enrichment because of
3 the large contributions from nonatmospheric sources of nitrogen and the influence of both
4 oxidized and reduced forms of nitrogen, particularly in large watersheds and coastal areas.
5 However, a strong relationship exists between atmospheric deposition of nitrogen and ecological
6 responses and effects in high alpine lakes in the Rocky Mountains because atmospheric
7 deposition is the only source of nitrogen to these systems. In addition, there is strong qualitative
8 evidence regarding the relationships between ecological responses, effects, and benefits
9 attributable to terrestrial nitrogen nutrient enrichment; however, the relative contributions of
10 oxidized versus reduced forms of nitrogen must also be taken into account. Based on the
11 scientific analyses presented by the ISA and the Risk and Exposure Assessment, negative
12 ecological effects due to aquatic and terrestrial acidification may be the most useful in terms of
13 creating a developing a secondary NO_x/SO_x NAAQS that reflects the ecological impacts due to
14 these pollutants.

15 A summary of the information presented by this Risk and Exposure Assessment that may
16 be useful for characterizing known or anticipated adverse effects to public welfare is shown in
17 **Table 7.3-1**. This information may be useful to inform decision makers about what levels of
18 protection might be appropriate to protect public welfare from known or anticipated adverse
19 impacts on ecosystems. Characterizing known or anticipated adverse effects to public welfare
20 from a policy perspective will be addressed in the policy assessment for this review.

1 **Table 7.3-1.** Summary of Information Assessed in the Risk and Exposure Assessment to Aid in Informing Policy Based on Welfare
2 Effects.

Exposure Pathway (Current Deposition Levels) (NADP/CMAQ 2002)	Affected Ecosystem (Case Study Locations)	Ecological Response (Targeted Effect)	Ecological Indicator	Ecological Effect	Ecosystem Service Affected
Adirondack Case Study Area: 10 kg N/ha/yr 9 kg S/ha/yr Shenandoah Case Study Area: 11 kg N/ha/yr 11 kg S/ha/yr	Adirondack Mountains, NY Blue Ridge Mountains and Shenandoah National Park, VA	Acidification in lakes and streams	Fish species richness, abundance, composition, ANC	Species losses of fish, phytoplankton, zooplankton; changed community composition, ecosystem structure, and function	Annual recreational freshwater fishing in New York State = more than 13 million days Approximately \$66.4 million in implied value to NY anglers from a zero- out of N+S deposition
Kane Experimental Forest Case Study Area: 14 kg N/ha/yr 210 kg S/ha/yr Hubbard Brook Experimental Forest Case Study Area: 8 kg N/ha/yr 7 kg S/ha/yr	Kane Experimental Forest (Allegheny Plateau, PA) Hubbard Brook Experimental Forest (White Mountains, NH)	Acidification of forest soils	Tree health Red spruce, sugar maple Bc/Al ratio	Decreased tree growth Increased susceptibility to stress, episodic dieback; changed community composition, ecosystem structure, and function	Provision of wood products (sugar maple) 900 million board feet timber production

Exposure Pathway (Current Deposition Levels) (NADP/CMAQ 2002)	Affected Ecosystem (Case Study Locations)	Ecological Response (Targeted Effect)	Ecological Indicator	Ecological Effect	Ecosystem Service Affected
<p>Potomac River/Potomac Estuary Case Study Area: 13 kg N/ha/yr</p> <p>Neuse River/Neuse River Estuary Case Study Area: 14 Kg N/ha/yr</p>	<p>Potomac River Basin, Chesapeake Bay</p> <p>Neuse River Basin, Pamlico Sound</p>	Nutrient enrichment in main stem river of an estuary	ASSETS EI	Habitat degradation, algal blooms, toxicity, hypoxia, anoxia, fish kills, decreases in biodiversity	Current saltwater recreational fishing 26.1 million activity days (North Carolina-Massachusetts)
<p>Coastal Sage Scrub from 3 to 10 kg N/ha/yr</p> <p>Mixed Conifer Forest (San Bernardino Mountains and Sierra Nevada Range): from 3 to 10 kg N/ha/yr</p>	<p>Southern California Coastal Sage Scrub</p> <p>Mixed Conifer Forest (San Bernardino Mountains and Sierra Nevada Mountains, CA)</p>	Nutrient enrichment in terrestrial ecosystems	Species composition	Species changes, nutrient enrichment of soil, changes in fire regime, changes in nutrient cycling	Annual benefits to California residents hunting, fishing, and wildlife viewing = approximately \$4.6 billion; state expenditures for fire suppression = \$300 million (2008)

1 **7.4 REFERENCES**

- 2 Aber J.D., C.L. Goodale, S.V. Ollinger, M.L. Smith, A.H. Magill, M.E. Martin, R.A. Hallett, and
3 J.L. Stoddard. 2003. Is nitrogen deposition altering the nitrogen status of northeastern
4 forests? *Bioscience* 53:375–389.
- 5 Allen, E.B. 2008. Personal communication from E.B. Allen, University of California, Riverside,
6 CA.
- 7 Baron, J.S. 2006. Hindcasting nitrogen deposition to determine an ecological critical load.
8 *Ecological Applications* 16(2):433–439.
- 9 Baron, J.S., H.M. Rueth, A.M. Wolfe, K.R. Nydick, E.J. Allstott, J.T. Minear, and B. Moraska.
10 2000. Ecosystem responses to nitrogen deposition in the Colorado Front Range.
11 *Ecosystems* 3:352–368.
- 12 Bytnerowicz, A., and M.E. Fenn. 1996. Nitrogen deposition in California forests: a review
13 *Environmental Pollution* 92(2):127–146.
- 14 CAL FIRE (California Department of Forestry and Fire Protection). 1996. California Fire Plan.
15 Available at http://cdfdata.fire.ca.gov/fire_er/fpp_planning_cafireplan.
- 16 CAL FIRE (California Department of Forestry and Fire Protection). 2008. CAL FIRE 2007
17 Wildland Fire Summary.
- 18 Clark, C.M., and D. Tilman. 2008. Loss of plant species after chronic low-level nitrogen
19 deposition to prairie grasslands. *Nature* 451:712–715.
- 20 Cordell, K., B. Leeworthy, G.T. Green, C. Betz, and B. Stephens. n.d. *The National Survey on*
21 *Recreation & the Environment*. Research Work Unit 4953. Athens, GA: Pioneering
22 Research on Changing Forest Values in the South and Nation USDA Forest Service
23 Southern Research Station. Available at www.srs.fs.fed.us/trends.

- 1 Egerton-Warburton, L.M., and E.B. Allen. 2000. Shifts in arbuscular mycorrhizal communities
2 along an anthropogenic nitrogen deposition gradient. *Ecological Applications* 10(2):484–
3 496.
- 4 Fenn, M.E., M.A. Poth, S.L. Schilling, and D.B. Grainger. 2000. Throughfall and fog deposition
5 of nitrogen and sulfur at an N-limited and N-saturated site in the San Bernardino
6 Mountains, southern California. *Canadian Journal of Forest Research* 30:1476–1488.
- 7 Fenn, M.E., J.W. Baron, E.B. Allen, H.M. Rueth, K.R. Nydick, L. Geiser, W.D. Bowen, J.O.
8 Sickman, T. Meixner, D.W. Johnson, and P. Neitlich. 2003. Ecological effects of
9 nitrogen deposition in the western United States. *Bioscience* 53(4):404–420.
- 10 Fenn, M.E., Huntington, T.G., McLaughlin, S.B., Eager, C., Gomez, A., and Cook, R.B. 2006.
11 Status of soil acidification in North America. *Journal of Forest Science* 52(special issue):
12 3–13.
- 13 Fenn, M.E., L. Geiser, R. Bachman, T.J. Blubaugh, and A. Bytnerowicz. 2007. Atmospheric
14 deposition inputs and effects on lichen chemistry and indicator species in the Columbia
15 River Gorge, USA. *Environmental Pollution* 146:77–91.
- 16 Fenn, M.E., S. Jovan, F. Yuan, L. Geiser, T. Meixner, and B.S. Gimeno. 2008. Empirical and
17 simulated critical loads for nitrogen deposition in California mixed conifer forests.
18 *Environmental Pollution* 155(3):492–511.
- 19 Grulke, N.E., C.P. Andersen, M.E. Fenn, and P.R. Miller, 1998. Ozone exposure and nitrogen
20 deposition lowers root biomass of ponderosa pine in the San Bernardino Mountains,
21 California. *Environmental Pollution* 103:63–73.
- 22 Grulke, N.E., C. Andersen, and W.E. Hogsett. 2001. Seasonal changes in above- and
23 belowground carbohydrate concentrations of ponderosa pine along a pollution gradient.
24 *Tree Physiology* 21:175–184.
- 25 Grulke, N.E., R.A. Minnich, T.D. Paine, S.J. Seybold, D. Chavez, M.E. Fenn, P.J. Riggan, and
26 A. Dunn. 2008. Air pollution increases forest susceptibility to wildfires: a case study for

- 1 the San Bernardino Mountains in southern California. In *Wild Land Fires and Air*
2 *Pollution*, 8. Edited by A. Bytnerowicz, M. Arbaugh, A. Riebau, and C. Andersen.
3 Burlington, MA: Elsevier.
- 4 Kaval, P., and J. Loomis. 2003. *Updated Outdoor Recreation Use Values With Emphasis On*
5 *National Park Recreation*. Final Report October 2003, under Cooperative Agreement CA
6 1200-99-009, Project number IMDE-02-0070.
- 7 Lien, L., G.G. Raddum, and A. Fjellheim. 1992. *Critical Loads of Acidity to Freshwater: Fish*
8 *and Invertebrates*. The Environmental Tolerance Levels Programme. Rep. No. 23/1992.
9 Norwegian Ministry of Environment, Oslo, Norway.
- 10 McNulty, S.G., E.C. Cohen, H. Li, and J.A. Moore-Myers. 2007. Estimates of critical acid loads
11 and exceedences for forest soils across the conterminous United States. *Environmental*
12 *Pollution* 149:281–292.
- 13 MEA (Millennium Ecosystem Assessment Board). 2005a. *Ecosystems and Human Well-being:*
14 *Current State and Trends, Volume 1*. Edited by R. Hassan, R. Scholes, and N. Ash.
15 Washington, DC: Island Press. Available at
16 <http://www.millenniumassessment.org/documents/document.766.aspx.pdf>.
- 17 Meixner, T., and M. Fenn. 2004. Biogeochemical budgets in a Mediterranean catchment with
18 high rates of atmospheric N deposition – Importance of scale and temporal asynchrony.
19 *Biogeochemistry* 70:331–356.
- 20 National Association of State Foresters (NASF). 2009. Quadrennial Fire Review
21 2009. Washington, DC: NASF. *Quadrennial Fire and Fuel Review Final Report 2009*.
22 National Wildfire Coordinating Group Executive Board January
- 23 NEG/ECP Forest Mapping Group (Conference of New England Governors and Eastern
24 Canadian Premiers Forest Mapping Group). 2001. *Protocol for Assessment and Mapping*
25 *of Forest Sensitivity to Atmospheric S and N Deposition: Acid Rain Action Plan – Action*
26 *Item 4: Forest Mapping Research Project*. Prepared by NEG/ECP Forest Mapping
27 Group. Available at

- 1 [http://www.nrs.fs.fed.us/clean_air_water/clean_water/critical_loads/local-](http://www.nrs.fs.fed.us/clean_air_water/clean_water/critical_loads/local-resources/docs/NEGECP_Forest_Sensitivity_Protocol_5_21_04.pdf)
2 [resources/docs/NEGECP_Forest_Sensitivity_Protocol_5_21_04.pdf](http://www.nrs.fs.fed.us/clean_air_water/clean_water/critical_loads/local-resources/docs/NEGECP_Forest_Sensitivity_Protocol_5_21_04.pdf).
- 3 Rueth, H.M., J.S. Baron, and E.J. Allstott. 2003. Responses of Engelmann spruce forests to
4 nitrogen fertilization in the Colorado Rocky Mountains. *Ecological Applications* 13:664–
5 673.
- 6 Stoddard, J., J.S. Kahl, F.A. Deviney, D.R. DeWalle, C.T. Driscoll, A.T. Herlihy, J.H. Kellogg,
7 P.S. Murdoch, J.R. Webb, and K.E. Webster. 2003. *Response of Surface Water Chemistry*
8 *to the Clean Air Act Amendments of 1990*. EPA 620/R-03.001. U.S. Environmental
9 Protection Agency, Office of Research and Development, National Health and
10 Environmental Effects Research Laboratory, Research Triangle Park, NC..
- 11 Sullivan, T.J., C.T. Driscoll, B.J. Cosby, I.J. Fernandez, A.T. Herlihy, J. Zhai, R. Stemberger,
12 K.U. Snyder, J.W. Sutherland, S.A. Nierzwicki-Bauer, C.W. Boylen, T.C. McDonnell,
13 and N.A. Nowicki. 2006. *Assessment of the Extent to Which Intensively-Studied Lakes*
14 *are Representative of the Adirondack Mountain Region*. Final report. New York State
15 Energy Research and Development Authority (NYSERDA), Albany, NY. Available at
16 <http://nysl.nysed.gov/uhtbin/cgisirsi/Qcwd6NzFby/NYSL/138650099/8/4298474>
17 (accessed November 1, 2007).
- 18 Takemoto, B.K., A. Bytnerowicz, and M.E. Fenn. 2001. Current and future effects of ozone and
19 atmospheric nitrogen deposition on California’s mixed conifer forests. *Forest Ecology*
20 *and Management* 144:159–173.
- 21 U.S. Department of the Interior, Fish and Wildlife Service, and U.S. Department of Commerce,
22 U.S. Census Bureau. 2007. *2006 National Survey of Fishing, Hunting, and Wildlife-*
23 *Associated Recreation*.
- 24 U.S. EPA (Environmental Protection Agency). 2006. *Ecological Benefits Assessment Strategic*
25 *Plan*. EPA-240-R-06-001. U.S. Environmental Protection Agency, Office of the
26 Administrator, Washington, DC. Available at <http://www.epa.gov/economics>.

- 1 U.S. EPA (Environmental Protection Agency). 2008. *Integrated Science Assessment (ISA) for*
2 *Oxides of Nitrogen and Sulfur–Ecological Criteria (Final Report)*. EPA/600/R-
3 08/082F. U.S. Environmental Protection Agency, National Center for Environmental
4 Assessment–RTP Division, Office of Research and Development, Research Triangle
5 Park, NC. Available at <http://cfpub.epa.gov/ncea/cfm/recordisplay.cfm?deid=201485>.
- 6 Watmough, S. A., Eimers, M. C., Aherne, J., Dillon, P. J. (2004) Climate effects on nitrate
7 export from forested catchments in south-central Ontario. *Environmental Science and*
8 *Technology* 38: 2383-2388.
- 9 Weiss, S.B. 1999. Cars, cows, and checkerspot butterflies: Nitrogen deposition and management
10 of nutrient-poor grasslands for a threatened species. *Conservation Biology* 13:1476–1486.
- 11 Wood, Y., T. Meixner, P.J. Shouse, and E.B. Allen. 2006. Altered Ecohydrologic response
12 drives native shrub loss under conditions of elevated N-deposition. *Journal of*
13 *Environmental Quality* 35:76–92.
- 14

United States
Environmental Protection
Agency

Office of Air Quality Planning and Standards
Health and Environmental Impacts Division
Research Triangle Park, NC

Publication No. EPA-452/P-09-004a
June 5, 2009
